THE FATE OF MERCURY IN MUNICIPAL SOLID WASTE LANDFILLS AND ITS POTENTIAL FOR VOLATILIZATION

By

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Mercury is conveyed into landfills primarily via batteries and a variety of other mercury-containing devices and lamps. Mercury has not been the focal point of metals research in landfills, but the first Phase of this study showed that mercury exists in the Alachua County Landfill at concentrations ranging from 32.8 ng Hg/g to greater than 16,800 ng Hg/g. However, over half of the samples had concentrations of 150 ng/g or less. While Hg concentrations in the landfill samples as well as compost samples (1.1 yard

waste to biosolids) from Palm Beach County were generally above background levels for surface soils in Florida, they were two to three orders of magnitude lower than clean-up goals currently used by the Florida Department of Environmental Protection and federal regulations governing land application of sewage sludge as described in 40 CFR Part 503.

Phase II of this study investigated the fate of mercury in simulated landfills and found that the bulk of the Hg added was found in the solid waste. Sixteen out of 18 leachate samples did not have detectable levels of Hg. The percentage of Hg volatilized during anaerobic digestion reanged from over 30% at the lowest Hg level (100 ng Hg per 60 g solid waste) to about 3% at the highest Hg level (2000ng Hg per 60 g solid waste. Evidence was obtained that forms of Hg other than elemental or divalent Hg were volatilized. These other forms were thought to be organic Hg compounds, quite possibly including dimethylmercury.

Sulfur-impregnated, activated charcoal was used to trap Hg volatilized during anaerobic digestion. A modification of EPA Method 7471 successfully recovered >98% of the Hg trapped by the charcoal.

This study has implicated landfills as a potential source of mercury to the atmosphere. Further research should focus on quantifying the amount of mercury being emitted into the atmosphere from landfills and identifying those stages during anaerobic digestion of MSW that Hg is most likely to volatilize.

CHAPTER 1
INTRODUCTION

The overall objective of this project was to determine the fate of mercury (Hg) in a municipal solid waste (MSW) landfill and to establish its potential for impacting atmospheric pollution. Mercury is one of the most toxic heavy metals in existence. It is present in the atmosphere, aquatic systems, terrestrial systems, and biota that exist within these systems. Mercury is unique in its ability to exist as a liquid at room temperature. It also exists in nature in gaseous, liquid, and solid phases.

√Studies have confirmed that anthropogenic activities have increased the input of Hg to the environment. It is estimated that Hg from these activities constitute about half of the Hg entering the environment (Fitzgerald and Clarkson, 1991). Pirrone et al. (1996) have stated that Hg emissions in developed countries increased at a rate of 4.5-5.5% per year until 1989 and have remained somewhat constant since. In developing countries, emissions have steadily climbed at a rate of 2.7-4.5% per year. Mercury is present in fossil fuels and to a lesser degree, coal. The vast amounts of fossil fuels and coal burned annually represent an important source of Hg released into the biosphere (Mitra, 1986).

Over the years, there has been interest in utilizing solid waste (SW) residues for land application. However, little or no data exist in the literature with respect to Hg levels in MSW landfills. In Florida an attempt is being made to set standards for heavy metals in these residues, and there is need for more data to allow for the establishment of threshold levels in SW. Currently, stabilized MSW residues are tested for heavy metals using USEPA 503 regulations that were developed for sewage sludge. Mercury ranks as number three on the EPA's toxic substances list, behind lead and arsenic. Since Hg is one of the most harmful heavy metals in the environment, the fate of Hg in MSW should be known. Successful completion of this research may assist in the establishment of sampling protocols and regulations specifically for Hg contained in SW residues.

Several Hg-containing components enter the SW stream on a daily basis and eventually make their way to MSW landfills. Fluorescent lights, Hg vapor lamps, arc lamps, mirror coating, amalgam, and electrical apparatus are important sources of Hg in MSW. Although laws were passed in 1988 prohibiting the disposal of batteries in landfills batteries no doubt still find their way into them (Bureau of Solid and Hazardous Waste, 1995, Tchobanoglous, 1993; Steinwachs, 1990).

Research has established that Hg binds strongly to organic matter. There is a significant proportion of organic material in SW, and it is expected that the majority of the Hg existing in a landfill will be bound to it. Bacteria commonly found in landfills influence the transformations of Hg that is bound to organic matter. Studies have confirmed that many of these bacteria contribute to methylmercury production. Thus, volatilization of dimethylmercury and elemental Hg from landfills could make a significant contribution to atmospheric Hg emissions. It has also been shown that inorganic mercury (Hg^{2}) can volatilize at the higher temperatures found during anaerobic digestion further contributing to atmospheric emissions.

Since there is need to understand the fate of Hg in a landfill, a study was conducted with the following objectives:

(1) Determine Hg concentrations in SW samples collected at

various depths over a five-year period from the Alachua County SW Landfill in Florida.

(2) Evaluate the potential for Hg present in landfills to be volatilized and released to the atmosphere.

These objectives were accomplished in two phases. In Phase I, total Hg levels in residues from the Alachua County landfill were determined. This landfill has been in existence since the early 1970's and has been the primary recipient of MSW from the city of Gainesville since then. Hundreds of residue samples from the Alachua County landfill had been collected and stored during previous studies (Miller et al., 1996; Miller et al., 1994). These samples were analyzed for total Hg to generate a large database for Hg in MSW residues from a landfill. The results for this landfill should be representative of landfills created by many other municipalities around the Southeastern United States.

In Phase II, the fate of Hg occurring in a landfill was studied using laboratory-scale anaerobic digester to which various amounts of Hg were added. In this phase, the distribution of Hg between leachate, solid, and gas phases was determined after a period of anaerobic digestion. Conditions occurring in these small-scale digesters are considered to be representative of those that occur in landfills similar to the one in Alachua County

CHAPTER 2
LITERATURE REVIEW

The earth's crust has been shown to contain low concentrations of mercury. Its main deposits occur in the form of cinnabar, or mercuric sulfide (HgS), which has an average mercury content of 0.1 to 0.4% (Mitra, 1986). In certain locations in the world such as Spain, Yugoslavia, Italy, Peru, Mexico and the American Continent, mercury is found in impregnated schist or slate and as geodes of liquid mercury. Mercury is also found in nature as the oxide and the selenide and in combination with a number of minerals such as quartz, dolomite, chalcedony, calcite, and pyrite. Mercury is present in fossil fuels, and to a lesser degree, coal. The vast amount of coal burned annually (between 1.4 x 10^{9} and 2.72 x 10^{9} g/yr) represents an important source of mercury released into the biosphere (Mitra, 1986).

A large quantity of mercury in the environment is derived from industrially produced mercury amounting to approximately 10,161 metric tons per year. Industries which are suspected as being responsible for the dispersion of mercury are MSW incinerators, medical waste incinerators, fossil fuel burning, chlor-alkali industries, mining and extraction of mercury from cinnabar, amalgamation, electrical equipment, paper pulp, fungicides, instrumentation, crematoria, degassing of latex paint, and cement manufacturing (Pirrone et al., 1996). Exposure to mercury in the home, hospital, or laboratory can occur by the release of mercury from broken thermometers (Stewart and Bettany, 1982; Mitra, 1986).

In the past, an estimated 90 718 kilograms of mercury was used in the USA each year in dentistry for the preparation of dental amalgam restorations (Mitra, 1986). In the early '90s, it was reported that the ingestion of elemental mercury (Hg^0) from dental amalgams caused renal damage in laboratory animals (Barkay, 1992). Kunkel et al. (1996) studied the fate of mercury in dental amalgams and determined that soluble mercury was never detected in a treatment plant headworks probably due to the fact that it became insoluble in the sludge floc. Mercury levels and discharge in wastewater from dental clinics in Denmark was investigated by Arenholt-Bindslev and Larsen (1996). Clinics without amalgam separators had a mean value of 270 mg Hg/dentist/day while those equipped with amalgam separators had a mean value of 35 mg Hg/dentist/day. Several hundred grams of mercury/clinic may be discharged with wastewater each year.

Wilhelm et al. (1996) studied biological monitoring of mercury vapor exposure in dental students by scalp hair analysis in comparison to blood and urine. It was concluded that hair may be used as an indicator of internal uptake of mercury provided that the hair was not externally exposed to mercury vapor.

Forms and Toxicity of Mercury

Mercury can be divided into three main categories. These are elemental mercury (Hg^0), inorganic mercury (Hg^{2+}, and its compounds) and organic mercury (for example methylmercury (CH_3Hg^+), phenylmercury ($C_6H_5Hg^+$), ethylmercury ($CH_3CH_2Hg^+$)). These affect human health in one way or another. 'Metallic mercury and inorganic mercury compounds generally attack the liver and kidney, but they normally do not remain in the body long enough (24 hours for mercuric mercury) to accumulate to serious levels (Hammond, 1971).' The excretion of inorganic mercury occurs through the kidneys, liver (as bile), intestinal mucosa, sweat glands, and salivary glands.

Inhalation has been shown to be the main route of exposure for mercury vapor. The lungs are able to absorb the metal with nearly 100 percent efficiency. Mercury vapor is considered hazardous at concentrations greater than 0.05 mg/m^3. Once mercury vapor gets into the bloodstream, it is oxidized to Hg^{2+} (Barkay, 1992). Methylmercury and mercury vapor pose the greatest threat to human health.

Methylmercury is the most toxic form of mercury. Microbes methylate inorganic mercury, generally under anaerobic conditions, to produce methylmercury (Mitra, 1986). Fish and shellfish provide the major source of intake by humans. Methylmercury is rapidly absorbed from the gastrointestinal tract of humans and attacks the central nervous system. It is only slowly excreted. The main route of excretion in humans is via the feces, in which the rate is about ten times higher than that in urine.

Poisoning from methylmercury is characterized by sensory disorders, concentric constriction of the visual fields, impairment of hearing, symptoms from the autonomic nervous system, decreasing physical coordination, lose of memory as well as "mental disturbances" often referred to as Minamata disease. In the most severe case in adults, specific anatomical areas of the brain are damaged. The damage is irreversible because neuronal cells are destroyed (Fitzgerald and Clarkson, 1991). Pregnant women are spared from poisoning because the methylmercury quickly crosses the placental barrier to accumulate in their unborn children instead, thus causing teratogenesis and damage of their central nervous systems (D'itri and D'itri, 1977).

Special precautions must be taken in the laboratory during the handling of mercury, especially the organic mercury forms. The death of the Dartmouth professor, Dr. Karen Wetterhahn, in 1997 highlights the fact that latex gloves do not adequately protect the skin from absorbing dimethylmercury.

The most notable methylmercury case on record occurred in Minamata, Japan during the 1950's. Initially, there were deaths of aquatic and terrestrial organisms. Then the impoverished fishermen and their families who ate large amounts of fish from Minamata Bay developed the strange symptoms mentioned above. It was concluded that these symptoms were caused by people eating fish and shellfish contaminated by methylmercury, the source of which was Hg that was discharged in the wastewater from the Chisso plant. This plant manufactured acetaldehyde and the resulting waste which contained inorganic mercury had been continuously dumped into Minamata Bay since the 1930's. In the sediment, the inorganic mercury was transformed to methylmercury and it eventually accumulated in the fish (Fujiki and Tajima, 1992; D'itri, 1991).

In the biosphere, elemental mercury has a high vapor pressure (Henry's constant, H=0.3) and a low aqueous solubility of 6 x 10^{-5} g/100 ml water at 25°C. Mercuric ion has a low vapor pressure ($H < 10^{-4}$) and a high aqueous solubility of 7.4 g/100 ml cold water (for $HgCl_2$). It forms covalent bonds, has an affinity for thiol groups, and tends to form strong bonds with inorganic and organic ligands. Organic mercury consists of highly stable C-Hg bonds, is very soluble in both water and hydrocarbons, and undergoes rapid transport through membranes. The vapor pressure of methylmercury (CH_3Hg^+) is quite low ($H < 10^{-7}$) but the vapor pressure of diethylmercury ($(CH_3)_2Hg$)) is high ($H < 0.3$) (Berkay, 1992).

Mercury has three oxidation states: 0, +1, and +2. Cationic mercury binds tightly to iron (Fe), aluminum (Al) and manganese oxides (MnO_2), hydroxides (OH), clay particles and silica. It also has a great affinity to sulfur (HgS), SH groups, and chlorine ($HgCl_2$).

Mercury in the Atmosphere

In the atmosphere, mercury may exist in the gaseous phase, the aqueous phase, and the solid particulate phase. Components of the gaseous phase are elemental mercury (Hg^0), mercuric chloride ($HgCl_2$), mercuric hydroxide ($Hg(OH)_2$), monoalkyl derivatives, such as methylmercuric chloride (CH_3HgCl), and dialkyl derivatives, such as dimethylmercury ($(CH_3)_2Hg$). The aqueous phase typically includes $HgCl_2$, $Hg(OH)_2$ and sulfites (SO_3); and in the solid particulate phase, there are mercuric oxide (HgO) and mercuric sulfide (HgS) (Seigneur et al. 1994, Schroeder et al. 1991, Nriagu and Davidson 1986). Concentrations of Hg species in the atmospheric environment are quite varied. Typical gas phase concentrations for Hg^0 range from 2-5 ng/m^3 and liquid phase concentrations range from 6-27 x 10^{3} ng/L. In the case of inorganic mercury (Hg^{2+}), the typical gas phase concentration is in the range 0.09-0.15 ng/m^3 and the typical liquid phase concentration is in the range 3.5-13.3 ng/L (Seigneur et al. 1994).

It is estimated that 95% of atmospheric Hg exists in the form Hg^0 and has an atmospheric residence time of 0.7-2.0

years (Gustin *et al.*, 1996; Petersen *et al.*, 1995; Nater and Grigal, 1992). Seigneur *et al.* (1994) have recently shown that global atmospheric budgets of Hg indicate a Hg^0 half-life on the order of one year.

Slemr and Langer (1992) have found increases in atmospheric Hg concentrations from 1970 to 1990 of about 1.5% per year in the Northern Hemisphere and about 1.2% per year in the Southern Hemisphere. Swain *et al.* (1992) have determined approximately a 2% increase in Hg deposition rates in Minnesota and Wisconsin.

Atmospheric mercury sources

Natural sources of atmospheric Hg include windblown dust, volcanogenic particles, forest wildfires, and seasalt. Nriagu (1989) stated that volcanoes and fumaroles are responsible for 50% of the Hg that is released naturally, and soil-derived dusts, forest fires, and seasalt sprays account for less than 10% of Hg released from natural sources. Natural degassing rates of Hg on regional or global scales have been estimated to range from 0.02 to 0.03 μg/m² h (Lindberg, 1986).

Annual global emissions of mercury to the atmosphere have been reported as 0.4 x 10^6 kg/yr from natural sources and 110 x 10^6 kg/yr from anthropogenic sources (Schroeder *et al*, 1992

Nriagu and Davidson, 1986). Major anthropogenic sources include fossil fuel combustion, particularly coal combustion, production of non-ferrous metals, refuse incineration (e.g. MSW incineration) and fuel wood combustion. Pacyna (1987) has implicated coal combustion as the largest single source of atmospheric Hg pollution, accounting for 58% of the anthropogenic input of Hg to the atmosphere. More than 90% of Hg in coal is released as Hg^0. Refuse incineration accounts for 33-40% of atmospheric mercuury pollution (Pirrone et al., 1996).

Atmospheric Species and Reactions

Many chemical species that are involved in atmospheric Hg chemistry. These will be listed followed by their typical concentrations in brackets: Hg (0.25-0.6 ppt); Br_2, HBr (20ppt); I_2, HI (0-3 ppt); HCl (0.01-2.0 ppb); O_3 (0.02-0.40 ppm), OH (0.04-0.4 ppt); NO_2 ($<=1$ ppb-0.5 ppm); SO_2 (0.003-0.1 ppm); H_2S (0.0-0.01 ppm); N_2O (300 ppb); NH_3 (0.015-100 ppb); H_2O_2 (<1ppb); O_2 (2.09×10^5 ppm); and Cl_2 (0.0-8.0 ppb) (Seigneur et al., 1994).

Two types of reactions occur in the atmosphere, namely gas-phase reactions and aqueous phase reactions. Most gas-phase reactions involve Hg^0 reacting with atmospheric species such as O_3, O_2, Cl_2, H_2S and H_2O_2.

Table 2-1 shows gas phase reactions of Hg in the atmosphere (Seigneur et al., 1994). Aqueous phase reactions occur in rainwater, fogwater, cloudwater, and other forms of precipitation. Table 2-2 shows aqueous phase reactions of Hg in the atmosphere.

Mercury in Aquatic Systems

Most aquatic system studies have focused on Hg in lakes (Angstrom et al., 1994; Lange et al., 1993; Winfrey and Rudd 1990, Steffan et al., 1988). Some authors have researched mercury in oceans and estuarine waters (Fabris et al., 1994; Langston, 1982; Mantoura et al., 1978; Lindberg and Harriss 1974). Mercury in rivers has been investigated by a few authors (Ebinghaus and Wilken, 1993; Bubb et al., 1991; Mantoura et al., 1978).

In freshwater, chlorides (Cl^-) and hydroxides (OH^-) are the main species existing, but sulfide (S^{2-}) may also be present. Elemental Hg and dimethylmercury (DMHg) are oxidized in the atmosphere to water-soluble species, such as Hg^{2+} and CH_3Hg^+, which are then deposited onto a wetland (Fitzgerald and Clarkson, 1991). When soluble MMHg enters the aquatic system, it is quickly accumulated by most aquatic biota (Clarkson et

Table 2-1 Gas Phase Reactions of Mercury

Reaction	Equilibrium or Rate Parameter (cm^3/molecule s)
(1) $Hg^0(g)+O_2(g) \dashrightarrow Hg(II)(g)$	$<8\ 0 \times 10^{19}$
(2) $Hg^0(g)+Cl_2(g) \dashrightarrow HgCl_2(g)$	$<4\ 1 \times 10^{16}$
(3) $Hg^0(g)+Br_2(g) \dashrightarrow HgBr_2(g)$	$<4\ 1 \times 10^{16}$
(4) $Hg^0(g)+I_2(g) \dashrightarrow HgI_2(g)$	$<2\ 7 \times 10^{16}$
(5) $Hg^0(g)+H_2O_2(g) \dashrightarrow Hg(OH)_2(g)$	$<4\ 1 \times 10^{16}$
(6) $Hg^0(g)+2NO_2(g) \dashrightarrow Hg(NO_2)_2(s,g)$	$3\ 3 \times 10^{35}$
(7) $Hg^0(g)+HCl(g) \dashrightarrow$ products	$1\ 0 \times 10^{19}$
(8) $2Hg^0(g)+O_2(g) \dashrightarrow 2HgO(s,g)$	$<1\ 0 \times 10^{33}$
(9) $Hg^0(g)+SO_2(g) \dashrightarrow$ products	$<6\ 0 \times 10^{1}$
(10) $Hg^0(g)+H_2S(g) \dashrightarrow$ products	$<6\ 0 \times 10^{17}$
(11) $Hg^0(g)+N_2O(g) \dashrightarrow$ products	$<2\ 0 \times 10^{17}$
(12) $Hg^0(g)+NH_3(g) \dashrightarrow$ products	$<1\ 0 \times 10^{17}$
(13) $Hg^0(g)+2HI(g) \dashrightarrow HgI_2(g)+H_2(g)$	$2\ 7 \times 10^{41}$
(14) $HgCl_2(g) \dashrightarrow$ products	slow
(15) $Hg(OH)_2(g) \dashrightarrow$ products	not available

Adapted from Seigneur et al (1994)

Table 2-2 Aqueous Phase Reactions of Mercury

Reaction	Equilibrium or Rate Parameter
1) Hg_2^{2+} <-->$Hg^0(aq)$+Hg^{2+}	2.9 x 10^{-9} M
(2) $Hg^0(aq)$+$O_3(aq)$-->Hg(II)(aq)+ $O_2(aq)$	4.7 x $10^7 M^{-1}s^{-1}$
(3) $Hg^0(aq)$+2HClO(aq)+$2H^+$+2e -->Hg^{2+}+$2Cl^-$+$2H_2O(l)$	not available
(4) $Hg^0(aq)$+Peracetic acid or m-chloro-peroxybenzoic acid(aq) -->Hg^{2+} or Hg	not available
(5) $Hg^0(aq)$+$H_2O_2(aq)$-->HgO(s)+ Hg^{2+}+$H_2O(l)$	6.0 $M^{-1}s^{-1}$
(6) Hg_2^{2+}+$O_3(aq)$-->Hg^{2+}	9.5 x $10^4 M^{-1}s^{-1}$
(7) Hg_2^{2+}+$H_2O_2(aq)$-->Hg^{2+}	<16 $M^{-1}s^{-1}$
(8) $Hg(SO_3)_2^{2-}$-->Hg^0	1.0 x 10^{-4} s^{-1}
(9) $HgSO_3(aq)$+ SO_3^{2-}<-->$Hg(SO_3)_2^{2-}$	2.5 x 10^{11} M^{-1}
(10) Hg^{2+}+SO_3^{2-}<-->$HgSO_3(aq)$	5.0x10^{12} M^{-1}
(11) $HgSO_3(aq)$-->$Hg^0(aq)$+SO_4^{2-}	0.6 s^{-1}
(12) $HgCl_2(aq)$ -->Hg^{2+}+$2Cl^-$	not available
(13) $Hg(OH)_2(aq)$-->$Hg^0(aq)$	not available
(14) $HgCl_2(s)$<-->$HgCl_2(aq)$	0.27 M
(15) $HgCl_2(aq)$<-->Hg^{2+}+$2Cl^-$	10^{-14} M^2
(16) $HgCl_2(aq)$+$2Cl^-$<-->$HgCl_4^{2-}$	70.8 M^{-2}

Table 2 2 Aqueous Phase Reactions of Mercury (continued)

Reaction	Equilibrium or Rate Parameter
(17) $Hg(OH)_2(s) <\!-\!-\!> Mg(OH)_2(aq)$	$3\ 5 \times 10^{4}$ M
(18) $Hg(OH)_2(aq) <\!-\!-\!> Hg^{2} + 2OH$	10^{22} M^2

Adapted from Seigneur _et al_ (1994)

al., 1984). Mercury bioaccumulates in aquatic organisms by three processes, namely: (1) from the water via respiration (e.g. over the gills) (2) by absorption of water from the body surface, and (3) by ingestion of food (D'Itri, 1991). The uptake of Hg through the aquatic food chain is an extremely important route of bioaccumulation. Mercury tends to bioaccumulate and biomagnify as it moves up a food chain.

Methylation

Methylation of Hg is a major step in biogeochemical cycling. Sediments are a part of the aquatic system, and are an active site for Hg methylation, being the primary sink for Hg released into the environment. In the sediment a variety of microbes transform the inorganic Hg into MMHg and DMHg.

Various studies have determined specific microorganisms that are responsible for the methylation of Hg in the environment. Wood *et al.* (1968) were the first to show that extracts of methanogenic bacteria (strict anaerobes) methylated Hg. Other studies implicated the bacteria *Neurospora* spp. (Landner, 1971), and *Clostridium cochlearium* (Yamada and Tonomura, 1972) in the methylation process, as well as a number of gram-negative and gram positive cocci isolated from river sediment (Hamdy and Noyes, 1975). Vonk and Sijpestein (1973) isolated *Pseudomonas* and *Bacillus* from soil, as well as culture strains of *Mycobacterium*, *Escherichia coli*, *Bacillus megaterium*, and fungi. Gilmour et al. (1992) determined that sulfate reducing bacteria were important in the methylation of Hg. Therefore, it can be stated that the bacteria which methylate Hg are facultative aerobes and anaerobes and sulfate-reducing bacteria.

Mercury that is buried in deeper layers of sediment is not available for methylation unless it is disturbed. Physical occurrences, such as shifting bottom currents, plus small worms, can stir the mercury-rich layers to a depth of two centimeters. Larger freshwater organisms, such as mussels can stir up the sediment to a depth of at least nine centimeters (D'itri and D'itri, 1977). More MMHg is present in water nearest to surface layers of sediment where facultative microbes usually live.

Three methylating coenzymes in biological systems are stated to participate in enzymatic methylation. These are (1) S-adenosylmethionine, (2) N^5-methyltetrahydrofolate derivatives, and (3) methylcobalamin (D'itri et al., 1978).

Shapiro and Schlenk (1965) and Ridley et al. (1977) discovered that S-adenosylmethionine and N^5-methyltetrahydrofolate could not transfer methyl groups to mercuric ions because they were only able to transfer a methyl group as CH_3^+, a carbonium ion. Methylcobalamin is able to transfer a methyl group to an inorganic mercuric ion. It can transfer groups as a carbanion (CH_3) and a methylradical (CH_3) to produce MMHg and DMHg under aerobic and anaerobic conditions (Wood, 1974).

When MMHg is taken up by fish, it moves to the red blood cells and then to fatty tissues where it may be retained for up to two years. Fish and shellfish provide the major source of intake by humans.

In humans MMHg is rapidly absorbed by the gastrointestinal tract and attacks the central nervous system. When MMHg combines with tissue it is stable and is very slowly degraded or excreted from the body (Mitra, 1986, Barkay, 1992). The main route of excretion in humans is via the feces, in which the rate is about ten times that in the urine (Mitra 1986). Studies with radioactive MMHg have demonstrated that it is retained in the human body with a half life of about 70 days. Thus, toxic amounts can be accumulated even with low dose rates (Hammond, 1971).

Demethylation

Methylmercury is slowly degraded because of the high stability of the carbon-mercury (C-Hg) bond. The low polarity coupled with low affinity of Hg to oxygen decreases the chances for hydrolytic cleavage of the strong bond. The means by which MHg is degraded is by the slow process of protolytic attack on the C-Hg bond. Clarkson et al. (1984) reported that the demethylation of MHg in the water phase is carried out by a variety of microorganisms in two enzymically mediated states: (1) hydrolase enzymes cleave the C-Hg bond releasing the methyl group, and (2) reductase enzymes convert ionic Hg to Hg^0 which can diffuse into the atmosphere. The equation below shows the transformation:

$$CH_3\text{-}Hg \xrightarrow{\text{hydrolase}} CH_4 + Hg^{2} \xrightarrow{\text{reductase}} Hg^0$$

The enzyme, organomercurial lyase, which is produced by bacteria in sediments, soils, and waters, can speed up this reaction 10^6 fold (Barkay, 1992). Thus, other authors have implicated a two step demethylation procedure involving cleaving of the C-Hg bond by organomercurial lyase to produce CH_4 and ionic Hg, followed by reduction of the ionic Hg to Hg^0

by the enzyme, mercuric reductase (Barkay, 1992; Oremland et al., 1991; Nakamura et al., 1990). The equation below shows this transformation:

$$CH_3\text{-}Hg \xrightarrow{\text{organomercurial lyase}} CH_4 + Hg^{2} \xrightarrow{\text{mercuric reductase}} Hg^{0}$$

Furthermore, it was stated that Hg volatilization has been found to be chromosomally encoded in specific bacterial genera, namely *Staphylococcus aureus* and *Bacillus* spp. (Nakamura et al., 1990).

Lakes

The primary source of Hg in lakes is from atmospheric deposition. Atmospheric deposition to lakes occurs mainly as inorganic Hg, even though there are small amounts of MHg (Winfrey and Rudd, 1990). In aerobic waters, Hg^{2} will complex with inorganic ligands such as chlorides and hydroxides, bind with dissolved organic carbon (DOC) or attach to particulate matter. Ionic Hg can also be microbially reduced to form Hg^{0}. In anaerobic zones, ionic Hg can be converted to MHg or complex with sulfides and precipitate as mercuric sulfide (HgS). Particle-bound Hg reacts with sulfides and is converted to insoluble HgS which then settles out of the water column onto the sediment. In the presence of hydrogen sulfide (H_2S), MHg will form methylmercuric sulfide $(CH_3Hg)_2S$ which decomposes to HgS and DMHg ($(CH_3)_2Hg$) which tends to volatilize (Winfrey and Rudd 1990; and Iverfeldt and Lindqvist, 1986).

/ Gilmour et al (1992) found the highest MHg concentrations near the sediment-water interface and in shallow sediments. Sulfate-reduction rates were also highest at the sediment water interface, thus confirming that sulfate reducing bacteria are important in the methylation of Hg.

/ Clarkson et al (1984) stated that inorganic Hg methylation rates by microorganisms are pH-dependent with the greatest amount of MHg being formed at pH levels lower than 6. Higher pHs yield DMHg which is volatilized. Bloom et al (1991) studied the impact of acidification on the MHg cycle of remote seepage lakes. Methylmercury concentrations were measured in the water of five pristine lakes which had a pH range of 4.6 to 7.2. In general, it was found that MHg in lake water tends to increase as pH decreases. Methylmercury partitioning was weakly related to pH.

* Miskimmin et al. (1992) studied the effects of pH on the Hg methylation and demethylation rates in lake water. The

results showed that a reduction in pH from 7.0 to 5.0 yielded large increases in net methylation rates at both low and high dissolved organic carbon (DOC) concentrations. Rates of microbial activity, which were represented by rates of respiration, had the least effect on net MHg production rates in the pH range 5.0 to 7.0.

Steffan et al. (1988) studied the effects of acidification on Hg methylation, demethylation, and volatilization in sediments from an acid susceptible lake. Sulfuric acid (H_2SO_4), hydrochloric acid (HCl) and nitric acid (HNO_3) were used to acidify the sediment. Methylation was inhibited over 65% when H_2SO_4 and HCl were used to reduce the pH from 6.5 to 4.5. There was almost complete inhibition of methylation when HNO_3 was used to bring the pH s to 5.5, 4.5 and 3.5. Demethylation was greatest at pH < 4.4, but was not affected by pH's between 4.4 and 8.0. Volatilization was less than 2% of methylation activity and was not significantly affected at the various pH levels.

Winfrey and Rudd (1990) determined that decreased pH stimulates MHg production at the sediment-water interface and probably in the aerobic water column. It was also shown that decreased pH also decreases loss of volatile Hg from lake water and increases Hg binding to particulates in water These factors enhance the bioavailability of Hg for methylation, therefore methylation rates may be increased In anoxic subsurface sediments, pH decreases the methylation rates, implying that formation in the water column and at the sediment water interface may be most significant in acidified lakes

Xun et al. (1987) reported that in the pH range 4 5 8 5 there was an inverse relationship between Hg methylation and pH Regnell (1994) did not find significant differences in methyl^{203}Hg in water at pH 5.8 and 6 6 when radiolabeled ^{203}HgCl$_2$ was added to water overlying sediment in freshwater systems The report also confirmed the findings of other authors that anoxic conditions enhanced MHg production in water (Olson and Cooper 1976).

Mantoura et al (1978) reported that in freshwater, more than 90% of Hg had been found to be complexed by humic materials.

Xu and Allard (1991) studied the effects of fulvic acid on the speciation and mobility of Hg in aqueous systems. It was determined that at pH levels below the point of zero charge, the humic substances adsorb and increase the uptake of trace metals from the solution phase. In the case of fulvic acid taking part in the adsorption of Hg on an oxide (alumina), the presence of the fulvic acid enhanced the Hg adsorption in the pH range of 2.5 - 9.5. In general, fulvic acid decreases Hg mobility under both acidic and basic conditions.

Mierle and Ingram (1991) investigated the role of humic substances in the mobilization of Hg from watersheds. Their results indicated that humic matter controls the solubility and watershed export of Hg deposited in precipitation. Minagawa and Takizawa (1980) determined very low levels of inorganic and organic Hg in natural waters by CVAAS after preconcentration on a chelating resin. In analyzing lake and river waters, it was found that 35-60% of the Hg present was in the form of organic compounds or in association with organic matter.

Hakanson (1980) established that higher MMHg levels were found in fish from waters with low productivity, low pH, and high water and sediment load. The reduction of sewage sludge in a lake was shown to increase MMHg content of fish because bioproductivity is decreased and pH lowered. The Hg cycle and fish in the Adirondack lakes was investigated. There was an observed pattern of increasing total Hg and MMHg with decreasing pH. Another correlation found was that lakes with higher dissolved organic carbon (DOC) had higher concentrations of both total Hg and MMHg. There were also positive correlations between fish Hg and MMHg concentrations in the water column.

In the case of wastewater, conventional methods for the removal of Hg^{2} include sulfide precipitation, ion exchange, alum and iron coagulation and adsorption on activated carbon. Namasivayam and Senthilkumar (1997) investigated the recycling of industrial solid waste for the removal of Hg^{2} by adsorption process. The report focused on the feasibility of Fe(III)/Cr(III) hydroxide for the adsorption of Hg^{2} from aqueous solution.

Terrestrial System

Soil

Mercury enters the soil via the disposal of sewage sludge, rainfall, dry fall-out from the atmosphere and the use of mercury based pesticides. A more recent addition would be land application of compost to condition the soil or fertilize crops or vegetation grown. Compost is solid waste that has undergone biological decomposition of the biodegradable organic matter under controlled mesophilic and thermophilic temperature conditions and has been stabilized to a degree which is potentially beneficial to plant growth. This stabilized material is used or sold for use as a soil amendment, artificial top soil, or other similar applications. The stabilized compost can easily and safely be stored, handled and used in an environmentally acceptable manner.

Mercury in the soil occurs in several forms. These are (1) dissolved (free ion or soluble complex), (2) non-specifically adsorbed (binding mostly due to electrostatic forces), (3) specifically adsorbed (strong binding due to covalent or coordinated forces); (4) chelated (bound to organic substances), and (5) precipitated (as sulfide, carbonate, hydroxide, phosphate, etc.) (Schuster, 1991).

Adsorption of Hg in soils is dependent on the chemical form of Hg, soil pH, amount and chemical nature of inorganic and organic soil colloids, type of exchangeable cations and redox potential (Adriano, 1986).

Coarse gravel has a lower capacity to bind Hg than finer soil materials, clay > silt > sand in the order of Hg complexation abilities. Farrah and Pickering (1978) investigated the uptake of Hg by three clay minerals and found illite > montmorillonite > kaolinite Reimers and Krenkel (1974) found similar results in that illite and montmorillonite had faster Hg uptake than kaolinite. Under acidic conditions, ion-exchange is the most important form of sorption to clay minerals, while Hg adsorption is assumed to occur mainly in the form of $Hg(OH)_2$ at higher pH values

Numerous studies have reported that Hg shows a great affinity for organic matter in soils peats and sediments (Newton et al., 1976; Wallace et al., 1982, Schuster 1991) The binding is strong, but reversible At low Hg concentrations, soil organic matter is responsible for most of the Hg sorption. At higher concentrations, mineral components take part in the sorption.

Results suggest that inorganic colloids contribute to the adsorption of organomercurials whereas inorganic Hg compounds tend to bind strongly to soil organic matter It was also stated that organic components were even more relevant in mercury adsorption at higher Hg concentrations This is because organic matter has a larger adsorption capacity for Hg than mineral colloids (Schuster, 1991)

Wilken (1992) stated that most of the water soluble Hg species in soil were complexed by organic material with a molecular weight greater than 500 daltons and particle sizes smaller than 1 2 µm Mercury is bound to humic matter by exchangeable cationic and non-cationic sites (Aggarwal and Desai 1980) About one-third of the total binding capacity of the soil humus is used for cation-exchange processes and about two-thirds of the available binding sites serve for metal complexation

Johansson et al. (1991) studied Hg in Swedish forest soils and waters and determined that the transport and distribution of Hg in forest soils is positively correlated to the transport of organic matter

The maximum sorption of Hg for several soil types occurs in the pH range 4.75-6.50 (Lodenius 1990). In acidic soil (pH < 4 5 to 5) organic material is the only sorbent for inorganic Hg In neutral soils iron oxides and clay minerals play a part in Hg sorption With decreasing pH the mobility of Hg increases in soils that are low in organic matter (Schuster, 1991)

Chlorides occur in all natural soil and water systems and it is regarded as one of the most mobile and persistent complexing agents for heavy metals (Schuster 1991 Adriano, 1986). Barrow and Cox (1992) investigated the effects of pH and chloride concentrations on the sorption of Hg. They concluded that in the absence of chloride, there were only infinitesimal effects of pH on sorption between 4 and 6. Sorption decreased at higher pH. At low pH the addition of chloride decreased sorption, but at high pH chloride additions had little effect on sorption.

Another study dealing with complex formation on Hg (II) adsorption by bentonite also found that chloride ions reduced Hg (II) adsorption, especially at low pH's. Maximum adsorption occurred in the pH range 4.5 to 5.5, regardless of initial Hg concentration.

The leaching of Hg from peat soils, such as those found in freshwater wetlands, decreases with decreasing pH (Lodenius, 1990). Lodenius et al (1987) studied the sorption and leaching of Hg in peat soil using small additions of labeled Hg in peat lysimeters. Most of the added Hg was bound to the uppper most layer of the peat columns. Additions of chloride and sterilant did not effect the leaching of Hg. When the peat soil was allowed to dry completely, the leaching of Hg was greatest. This may have been the result of Hg that was attached to suspended organic material passing quickly through the cracks that had formed in the soil. Also greater amounts of artificial rainfall resulted in increasing amounts of Hg being leached out.

Low organic matter acidic soils increase Hg mobility, and removal by leaching is more likely in acidic soils. Fang (1981) studied the sorption of Hg vapor in dry and moist soil columns. He found increased sorption with increasing soil moisture until a maximum was achieved near the maximal water holding capacity. Schnitzer and Kerndorff (1981) determined that humic substances tend to increase the solubility of Hg by the formation of water soluble complexes.

Comparing peat to sandy soils, the retention of mercury was much stronger and the volatilization smaller in peat than in sandy soils due to the tight bonding of Hg with organic matter (MacLean, 1974). In mineral soils, where Hg adsorption is dominated by iron oxides and clay minerals, there is promotion of evaporation as Hg^0. Humic acid enhances the reduction of Hg^{2+} to Hg^0 and thereby increases volatilization losses to the atmosphere (Rogers, 1977; Lia _et al._, 1982).

Bacteria and other microorganisms play a major part in Hg volatilization. They are able to reduce Hg^{2+} to Hg^0, which is volatile. They are also able to methylate Hg to DMHg, which is volatile, under alkaline conditions.

Some bacteria adsorb Hg on the outside of their cell walls where it converts directly into a vapor. *Escherichia coli* absorb Hg into their systems, then the Hg^{2+} may combine with the cytoplasm and convert into another compound or the volatile Hg^0. *Pseudomonas aeruginosa proteus* and at least two other microorganisms are known to convert Hg^{2+} into Hg^0 (D'itri and D'itri, 1977).

Mercury volatilization tends to increase with increasing temperature. Rogers and MacFarlane (1978) evaluated the volatilization rates of Hg in clay and sand and determined that the volatilization rate of Hg from clay was greater at higher temperatures, but less of the total Hg was volatilized. Increasing Hg concentration resulted in an increased volatilization of Hg from the sand and clay. Autoclaving bother soils drastically decreased the volatilization rates. After inoculation of the sterile soils with non-sterile soils, volatilization rates increased, thus indicating that it was microbially mediated. Another study found Hg to be translocated in soil horizons, and in some cases, lost from surface soil. This was attributed to evaporation to the atmosphere (Dudas and Pawluk, 1976).

Sediments

Accumulation rates for Hg in sediments have been estimated at between 10 and 50 μg Hg/m^{2} y (Andersson et al., 1990). Sediments are the primary sink for Hg released into the environment. Sediments consist of an oxidized zone and a reduced zone. In the oxidized zone, particulate Hg is desorbed to release Hg^{2} and methylated biotically and abiotically to produce MMHg (Zhang and Planas, 1994). In the oxidized zone, Hg^{2} can be readsorbed to form particulate mercury. The MMHg and particulate-Hg can be deposited into the reduced zone of sediments. In this zone, Hg^{2} is reduced to Hg and ultimately Hg^{0} which is then volatilized. Microbes aid in this reduction. In the reduced zone, ionic or Hg^{2} can react with sulfides that are present to form HgS. Methylmercury in this zone also reacts with sulfides (S^{2}) to form DMHg and HgS (Mitra, 1986). Mercury reactions that take place in the oxidized and reduced zones of sediment are given below;

(a) Oxidized zone reactions

(1) $$\text{particulate-Hg}^{2} \underset{\text{(adsorption)}}{\overset{\text{(desorption)}}{\rightleftharpoons}} \text{Hg}^{2}$$

(3) $$Hg^{2+} \xrightarrow{\text{(methylation)}} CH_3Hg^{+}$$

(b) Reduced zone reactions:

(1) $$Hg^{2+} \xrightarrow{\text{(reduction)}} Hg^{+} \xrightarrow{\text{(reduction)}} Hg^{0}$$

(2) $$Hg^{2+} \xrightarrow{+S^{2-}} HgS$$

(3) $$CH_3Hg \xrightarrow{+S^{2-}} (CH_3)_2Hg + HgS$$

Methanogens, along with sulfate reducers have been implicated in the methylation of Hg (Adriano, 1986; Wood 1972). Oremland *et al.* (1991) investigated the involvement of methanogens and sulfate reducers in oxidative demethylation. Under anaerobic conditions, results with inhibitors showed partial involvement of both sulfate reducers and methanogens. Sulfate reducers dominate estuarine sediments. Products of anaerobic demethylation were mainly carbon dioxide (CO_2) and methane (CH_4). Methane was the only product resulting from aerobic demethylation in estuarine sediments suggesting that the procedure for demethylation followed the organomercurial pathway. Final results of this study led to the conclusion that both aerobes and anaerobes demethylate Hg in sediments but either group may dominate in a specific sediment type.

An earlier study by Spanger *et al.* (1973) looked at MMHg and inorganic Hg in lake sediments. Bacterial isolates rapidly degraded MMHg to CH_4 and Hg^0. Heaton and Laitinen (1974) investigated the electrochemical reduction of MMHg. This occurs in two one-electron steps with the first electron resulting in the formation of a methylmercuric radical on the electrode and the second electron resulting in the reduction of methylmercuric compound to CH_4 and Hg^0. This also confirms the role played by methanogens in Hg methylation. Langston (1982) showed that humic and fulvic acids provided favorable binding sites that accounted for 4 to 32% of the total Hg measured in surface sediments. Evans *et al.* (1984) and Revis *et al.* (1990) found that the majority of extractable Hg was in the humic-fulvic acid and organic-sulfide fractions. Abiological methylation of Hg by humic acid has been confirmed in vitro (Nagase *et al.*, 1984).

Rekolainen *et al.* (1986) reported on the effect of airborne Hg and peatland drainage on sediment Hg contents in some Finnish forest lakes. The sediment Hg content correlated directly with the organic matter content of the sediment and the pH of the water

Di-Giulio and Ryan (1987) studied Hg in soils sediments and clams from a North Carolina peatland After selective extractions of peat and sediment samples, it was concluded that the majority of Hg was associated with organic matter associated fractions, particularly humic/fulvic bound and organic-sulfide bound

An important effect of pH is to mobilize MMHg sorbed on sediments A reduction in water pH shifts MMHg from the sediments to the water phase, regardless of the type of sediment (Miller and Akagi 1979)

Duarte et al (1991) studied Hg desorption from contaminated sediments Results showed that the amount of Hg desorbed from the sediment was inversely correlated with pH and ionic strength The effect of pH on the leaching of Hg from contaminated sediments was greatest at pH values less than 7 Concerning ionic strength the most Hg leaching occurred at ionic strength values less than 0 4 mol/dm^3

In anaerobic environments, such as sediments, or under mildly reducing conditions, there is a tendency for Hg to be precipitated as the sulfide (HgS) Mercuric sulfide is very insoluble (log K_s = -50.02 at 50°C) (IUPAC, 1982), but can be transformed under low Eh and high pH conditions to the soluble form, HgS_2^{2-}, or to Hg^0 (Barkay, 1992, Wilken 1992, Schuster, 1991). The insolubility of HgS makes it resistant to methylation, but under aerobic conditions, the sulfur in HgS may be oxidized to sulfate, after which the Hg^{2+} can undergo methylation (Adriano 1986).

Weber (1993) stated that sulfate-reducing bacteria contribute to MMHg production. A low sulfate concentration was needed for sulfate reducing bacteria to produce MMHg. Kerry *et al.* (1991) determined that methylation of Hg in sediments of an acid stressed lake was mainly due to the activity of sulfate reducing bacteria. The methylation rate did not correlate with the concentration of sulfate in the system. They speculated that formation of insoluble HgS reduced Hg availability, thus reducing MMHg production.

Revis *et al.* (1991) investigated the immobilization of Hg in soil, sediment, sludge, and water by sulfate-reducing bacteria. It was determined that the addition of calcium sulfate ($CaSO_4$) which slowly moves to the sulfur reducing bacteria, producing H_2S and eventually producing HgS, prevented the transformation of Hg to organic methylated forms

Gilmour et al (1992) reported that additions of sulfate to anoxic sediments yielded an increased microbial production of MMHg from added inorganic Hg. There were positive correlations between sediment depth profiles of bacterial sulfate reduction and Hg methylation. It was also noted that specific inhibition of sulfate-reducing bacteria blocked MMHg production at all depths in the sediment profile Sulfate-reducing rates and MMHg concentrations were highest near the sediment-water interface and in shallow sediments These results show that sulfate reducing bacteria are definite mediators of Hg methylation

Munthe et al (1991) investigated the aqueous reduction of Hg^{2} by sulfite This involved the formation of an unstable intermediate $HgSO_3$ which underwent decomposition to produce Hg , which in turn was reduced to Hg^{0} The concentration of sulfite inversely determined the overall rate of the reaction

Analytical Methods

Atmosphere

Lindberg et al (1992) investigated atmosphere exchange of Hg in a forest In this report Hg was collected in a series of gold traps, wet digested and then analyzed by long-path flameless atomic absorption. The detection limit for Hg in air ranged from 0.01 to 0.5 ng/m^3.

Licata et al. (1994) reviewed the testing of Hg in flue gases both in the USA and in Germany. In the USA, the most common method of analysis was CVAAS with $SnCl_2$ in a HCl solution as the reducing agent. In Germany, the most common method of analysis was also CVAAS but with $NaBH_4$ as the reducing agent.

Prestbo and Bloom (1995) investigated a Hg speciation adsorption (MESA) method for combustion flue gas. The sampling system for gas phase Hg species utilized a series of heated, solid phase adsorbent traps. Mercury (II) and monomethylmercury (MMHg) were the flue gas oxidized species that were adsorbed by a potassium chloride (KCl) impregnated soda lime sorbent. An iodated carbon sorbent was used to collect Hg^0 after passing through the KCl/soda lime sorbent. CVAFS was used for final determination of Hg from these sorbents.

Chu and Porcella (1995) investigated Hg stack emissions from US electric utility power plants. It was determined that Hg emissions from total electric utilities was on the order of

40 tons/yr. It was also determined that Hg emissions were not consistently captured by conventional air pollution control technologies including fabric filters, electrostatic precipitators, and flue gas desulfurization systems. The Hg in the emissions was analyzed by instrument neutron activation analysis (INAA). The MESA method was also used in this study, but neither this method nor the INAA was validated for Hg speciation.

Balogh and Liang (1995) investigated Hg pathways in municipal wastewater treatment plants that used incineration to dispose of the solid material. Iodated carbon traps were used to collect Hg in the incinerator exhaust gas. Subsequently, the traps were digested with acids and final analysis was done by CVAFS.

Keeler *et al.* (1995) studied particulate Hg in the atmosphere. The analytical technique performed on Hg(p) extracted from glass fiber and other types of filters from glass fiber: dual-amalgamation preconcentration and CVAFS detection. Teflon filters have been detected by instrumental neutron activation analysis (INAA).

Aquatic Systems

Minagawa and Takizawa (1980) determined very low levels of inorganic and organic Hg in natural waters by CVAAS after preconcentration on a chelating resin. A column of dithiocarbamate-treated resin was used to simultaneously collect inorganic and organic Hg at ng/L concentrations and subsequently quantitatively eluted with slightly acidic thiourea solution. Alkaline $SnCl_2$ solution is used to reduce inorganic Hg to Hg vapor. Mercury vapor was generated from inorganic and organic Hg with a $CdCl_2$-$SnCl_2$ solution. The range of determination was 0.2-5,000 ppt for 20 L water samples.

Determination of inorganic and organic Hg compounds by high performance liquid chromatography (HPLC)-inductively coupled plasma (ICP) emission spectrometry with cold vapor generation was investigated by Krull and Bushee (1986). It was not necessary to derivatize samples before analysis by HPLC. The conventional propylene spray chamber of the ICP was replaced by an all glass chamber. Detection limits ranged from 32 to 62 ppb of Hg for four Hg compounds. This represented three to four orders of magnitude enhancement over detection limits without cold vapor generation.

Xiankun et al. (1990) investigated the effect of the Huanghe river runoff on the occurrence, transportation, and speciation of Hg in the Huanghe Estuary and the adjacent sea. Mercury content in 150 mL filtered water samples was determined using two stage gold amalgamation coupled with CVAAS.

Lee and Iverfeldt (1991) measured Hg in run-off, lake and rain waters using an oxidative treatment with BrCl prior to reduction by $SnCl_2$. A gold trap was used to preconcentrate the Hg after its volatilization from a quartz glass reduction vessel. Nitrogen gas was used for the purging. A double amalgamation helium dc-plasma atomic emission method was used for analysis of the gold traps.

Baeyens and Belgium (1992) determined total Hg by acidification of the sample to pH 1, pretreatment with a strong oxidant, BrCl, followed by reduction of the BrCl with NH_2OH-HCl prior to $SnCl_2$ reduction, air purging of Hg^0 and collection on a Au column. Atomic fluorescence was used for analysis of these samples.

Smith (1993) determined natural levels of Hg in water samples by preconcentration onto gold traps followed by electrothermal heating and purging of the traps with argon directly into the torch of the ICPMS The detection limit was 0 2 ng/L using a 200 mL sample.

Garcia et al (1994) determined inorganic and MMHg in sea-water by an online preconcentration, solvent extraction and total Hg determination by CVAAS A detection limit of 16 ng/L of Hg was observed for sample volumes of 25 mL.

Bloom et al (1995) reported on the results of the international aqueous Hg speciation intercomparison exercise. Twenty three labs were involved with 18 utilizing BrCl oxidation gold trapping and CVAFS for total Hg. Four other labs used CVAAS or wet chemistry and the results were similar. Sixteen labs provided MMHg results, but 15 of them used various combinations of aqueous phase ethylation GC separation, and CVAFS detection

Emteborg et al (1995) used a dithiocarbamate resin for the sampling and determination of Hg species in humic-rich natural waters Filtration was used to collect the dithiocarbamate resin and transferred to a column which was placed into a closed flow injection system where acidic thiourea solution was used to elute the Hg species The separation was performed using a gas chromatograph equipped with a non-polar capillary column and detection utilized atomic emission spectrometry at 253.7 nm after excitation in a microwave-induced helium plasma.

Paquette and Helz (1995) reported on the solubility of cinnabar (Red HgS) and implications for Hg speciation in sulfidic waters. In this paper, the Hg analysis was performed by CVAAS on a homemade stannous chloride reduction, gold amalgamation apparatus attached to a Perkin-Elmer Model 2380 Spectrophotometer. A detection limit of 0.3 ppb Hg was noted.

Saouter et al. (1995) developed and field-validated a microcosm to simulate the Hg in a contaminated pond. The water samples were treated and preserved with BrCl and total Hg determination was accomplished using CVAFS.

Specific analytical techniques for organic Hg in aqueous samples have also been investigated. One of the earlier papers by Fitzgerald and Lyons (1973) dealt with trapping the Hg after its reduction while purging with nitrogen gas and concentrating it on a packed column immersed in a liquid nitrogen bath. After the completion of purging, the column was removed from the cold trap, heated, and the gas phase concentration of the eluted Hg was measured on a Coleman Hg analyzer (MAS-50). A detection limit of 0.0017 μg/L was obtained.

Schintu *et al.* (1987) developed a practical isolation technique for MMHg in natural waters. Mercury compounds were extracted quantitatively from six different sources of water with 5 mL of a 50 ppm dithizone-chloroform solution. This method provided a high recovery for both organic as well as inorganic Hg from an aqueous medium, prior to their determination by gold trap CVAAS.

Bloom and Watras (1989) developed a technique to quantify MMHg and DMHg in several rainfall and lake water samples by aqueous phase ethylation to the volatile dialkyl analogs followed by cryogenic gas chromatographic separation. Mercury-specific detection by CVAFS provided a 0.1 pg as Hg limit.

Lansens *et al.* (1990) determined MMHg in natural waters by headspace (HS) gas chromatography (GC) with microwave induced plasma (MIP) detection after preconcentration on a resin containing dithiocarbamate groups. The chelating resin, Sumilate Q-10, showed a high affinity for both organic and inorganic Hg.

Lee and Hultberg (1989) determined MMHg in some Swedish surface waters. Methylmercury was preconcentrated from 10-20 L of water on a sulfhydryl cotton fiber (SCF) adsorbent packed in a column and eluted with a small volume of 2M hydrochloric acid (HCl). The eluate was extracted with benzene and then analyzed for MMHg with the GC/ECD method.

Rapsomanikis and Craig (1991) investigated the speciation of Hg and MHg compounds in aqueous samples by gas chromatography-atomic absorption spectrometry after ethylation with sodium tetraethylborate. The absolute detection limit for CH_3HgCl was 167 pg.

Barzanini *et al.* (1992) simultaneously determined methyl, phenyl, ethyl, and inorganic Hg by CVAAS with on line chromatographic separation. Reversed-phase chromatography on an ODS column and elution with an acetonitrile water ammonium tetramethylenedithiocarbamate buffered mixture was investigated with or without ammonium tetramethylenedithiocarbamate precomplexation. The detection limits for methyl, ethyl, phenyl and inorganic Hg were 100 ng/mL, 50 ng/mL, 300 ng/mL, and 8 ng/mL respectively with direct injection (100 μL sample). For the on-line preconcentration procedure the detection limits were 0.5 ng/mL, 0.09 ng/mL, 0.5 ng/mL and 0.015 ng/mL respectively in a 100 mL sample.

Quevauviller et al. (1992) studied the occurrence of methylated tin and DMHg compounds in a mangrove core from Sepetiba Bay, Brazil. Mercury compounds were determined by derivatization with $NaBH_4$, cryogenic trapping in a chromatographic column and using an electrothermally heated quartz furnace by AA using an EDL source for final detection.

Gilmour and Bloom (1995) reported on a case study of Hg and MHg dynamics in a Hg-contaminated municipal wastewater treatment plant. Methylmercury was determined after distillation by ethylation, isothermal GC and CVAF detection.

Filippelli et al. (1992) investigated MMHg determination as volatile MHg hydride by purge and trap gas chromatography in line with fourier transform infrared spectroscopy. The detection limit of this method was 0.15 μg.

Table 2.3 provides a summary of the various analytical methods that the different authors used for the determination of mercury in aqueous samples.

Terrestrial System

Revis et al. (1990) determined MMHg in soil by developing a method for assessing acceptable limits. Water, acid copper sulfate ($CuSO_4$), sodium bromide (NaBr), and toluene were all added to a soil sample. The toluene phase was collected and

Table 2-2. An overview of the analytical methods for mercury in aqueous samples.

Analytical Methods	Species	Sources
Gold Trap CVAAS	Hg(Tot)	Xiankun et al (1990)
GC-HS-MIP	CH_3Hg	Lansens et al (1990)
GC-ECD	CH_3Hg	Lee & Hultberg(1990)
He dc plasma AED	Hg(Tot)	Lee & Iverfeldt(1991)
GC-QFAAS	CH_3Hg	Rapsomanikis & Craig (1991)
Gold Trap CVAFS	Hg(Tot)	Baeyens (1992)
GC-CVAAS	CH_3Hg	Serzanini et al. (1992)
Gold Trap with ICPMS	Hg(Tot)	Smith (1993)
CVAAS	Hg(Tot), CH_3Hg	Garcia et al. (1994)
CVAFS	Hg(Tot)	Saouter et al. (1995)
GC-CVAFS	Hg(II), CH_3Hg	Gilmour & Bloom(1995)
Gold Trap CVAAS	Hg(Tot)	Paquette & Helz (1995)

GC = gas chromatography
CVAAS = cold vapor atomic absorption spectrometry
CVAFS = cold vapor atomic fluoresence spectrometry
GC-HS MIP = headspace gas chromatography with microwave induced plasma detection
QFAAS = quartz furnace atomic absorption spectrometry
ICPMS = inductively coupled plasma mass spectrometry
AED = atomic emission detector
ECD = electron capture detector

mixed with an equal volume of sodium thiosulfate ($Na_2S_2O_3$) in ethanol. Potassium iodide (KI) and benzene were eventually added to the previous mixture. The benzene phase, which contains the MMHg was then analysed on the GC. The detection limit for MMHg was 30 ppb.

Hempel et al. (1992) determined organic Hg species in soils by high-performance liquid chromatography (HPLC) with ultraviolet (UV) detection. The simultaneous separation and quantification of nine organic Hg compounds was performed. This separation was done on octadecylsilane columns by gradient elution with a methanol-water mixture ranging from 30 to 50% v/v. The detection limits for the various compounds were in the range 70-95.1 $\mu g/dm^3$.

Evans et al. (1984) determined organic Hg in peat, sediment, and biological samples. The sediment aspect involved the use of acetone as an extractant, KBr and $CuSO_4$ in sulfuric acid and finally toluene as the last extractant. Mercury analysis was done using a graphite furnace AAS with the detection limit for MMHg set at 25 ng/g.

Mikac and Picer (1985) investigated Hg distribution in a polluted marine area. The methylmercury determination in the sediment samples involved HCl hydrolysis of the wet sample and extraction of CH_3HgCl into benzene. This extract was then dried with Na_2SO_4 and analyzed by GC/ECD.

Sakamoto et al. (1992) reported on the differential determination of organic Hg, Hg (II) oxide, and Hg (II) sulfide in sediments by CVAAS. All of these forms were removed from sediment by extracting with a solvent such as chloroform followed by a thiosulfate addition and subsequent analysis by CVAAS after digestion.

Engstrom et al. (1994) studied atmospheric Hg deposition to lakes and watersheds. Sediment cores were obtained from various systems and the samples were digested with a strong acid permanganate persulfate digestion technique. CVAAS was used for the total Hg analysis of these samples.

Zhang and Planas (1994) reported on biotic and abiotic Hg methylation and demethylation in sediments. Methylmercury was extracted from sediment samples using $CuSO_4/NaBr/H_2SO_4$ and toluene and analysis was performed by GC.

Rood et al. (1995) investigated Hg accumulation trends in Florida Everglades and Savannas Marsh flooded soils. The Hg in the soil and sediment samples underwent a strong digestion using an acid-permanganate persulfate combination with final analysis by CVAAS.

Municipal Solid Waste

Municipal solid waste consists of community refuse, including garbage, rubbish and trash (Nathanson, 1986). Average composition of the MSW stream in the USA includes paper and paperboard (41.0%), glass (8.2%), metals (8.7%), plastics (6.5%), rubber and leather (8.1%), food wastes (7.9%), yard wastes (17.9%) and other miscellaneous materials (1.6%) (Tchobanoglous, 1993; Concern, Inc., 1988). Predictions of increases in MSW (MSW) up to the year 2000 indicate increases in paper and paperboard, rubber and leather goods, plastics, yard waste, and metals. There are decreasing trends for food waste and glass (Tchobanoglous, 1993; Concern, Inc., 1988). In 1987-1988, 80% of MSW generated in the USA was landfilled, 10% was incinerated and 10% was recycled. Other amounts were dumped illegally on land and in the ocean. MSW is regulated under Subtitle D of the Resource Conservation and Recovery Act (RCRA) in 40 CFR Part 257 (USEPA, 1988).

In Florida, for the 12 month period ending June 30, 1993, 21.5 million tons of MSW were generated (up from 20.3 million tons in 1992). Of that amount, 9.9 million tons were landfilled (down from 10.4 million tons in 1992), 6.6 million tons were recycled (up from 5.4 million tons in 1992) and 5 million tons were combusted (up from 4.5 million tons in 1992) (Florida Department of Commerce, 1994). This is an excellent indication, at least within the State of Florida, that landfilling is decreasing and recycling is on the rise.

The MSW stream contains hazardous household and commercial toxic products of which heavy metals are included. The presence and mobility of heavy metals in MSW have been a source of concern for quite some time. MSW is landfilled, incinerated, or composted and the environmental issues with heavy metals focus on groundwater contamination, air pollution, and uptake by crops and other animals, and eventually humans. Hazardous wastes are a subset of MSW and can cause serious illness, injury, or death. They also present a problem to the environment if improperly transported or disposed (Nathanson, 1986). Heavy metals can be classified as hazardous wastes due to their various characteristics as stated in 40 CFR 261 subpart C which lists the four characteristics of hazardous waste as ignitability, corrosivity, reactivity, and extraction procedures (EP) toxicity. On the other hand, there are many items that are excluded from the definition of hazardous wastes in EPA regulations, of which the first item listed is household waste Many heavy metals found in MSW are actually products of household waste (Congress of the United States 1983)

Types of Heavy Metals

Many different types of heavy metals are found in the MSW stream it is important to note that aluminum and tin are not considered when referring to heavy metals included in the classification of heavy metals that exist are antimony (Sb) arsenic (As) cadmium (Cd), copper (Cu), chromium (Cr) iron (Fe) lead (Pb) manganese (Mn), nickel (Ni), zinc (Zn) mercury (Hg), selenium (Se), silver (Ag) and tin (Sn) Maximum concentration levels (in mg/l) as determined with the TCLP method for some of the heavy metals are 5 0 for arsenic 100.0 for barium. 1.0 for cadmium 5.0 for chromium 5 0 for lead, 0.2 for mercury, 1 0 for selenium and 5 0 for silver (Congress of the United States 1983)

Cadmium copper, nickel, zinc and molybdenum have been identified as the heavy metals with the greatest potential to accumulate in plants Cadmium can easily accumulate in crops at concentrations which could increase the dietary intake of the metal without causing crop phytotoxicity. Lead, mercury, arsenic and selenium are not considered to be much of a threat to plants because they have low solubility in slightly acid or neutral, well-aerated soils (Louisiana Cooperative Extension Service, 1991).

Sources

The major sources of heavy metals in MSW are lead acid batteries, household batteries (auto and flashlight), consumer electronics, plastics, used motor oil and transmission fluid, magazine ink, stain, varnish, and sealant (Bretz, 1990; USEPA, 1988). Fluorescent lamps, high pressure sodium lamps, and high intensity discharge lamps contain mercury. To date, the lamps still exceed the regulatory level of 0.2 mg/L for mercury and also exceed the regulatory level of 5.0 mg/L for lead when TCLP are performed (USACHPPM, 1997). Outdated light bulbs contain around 20 to 80 milligrams of liquid mercury. The breaking of one bulb can poison 50 cubic meters of breathing air (Kovalov, 1994).

Household batteries are major contributors of heavy metals in the solid waste stream. Each year, over two billion batteries are deposited in SW facilities in the United States (USEPA, 1997). Therefore, in a city of 500,000 people, almost 9,000 pounds of Hg would potentially enter the air, earth, and water (Jade Mountain, Inc., 1997). The different types of battery systems may contain Hg, cadmium, nickel, zinc, manganese, and lithium. Although each of these metals have negative health and environmental effects, Hg and cadmium are of greatest concern. It has been estimated that up to half of the Hg that is utilized in the United States is used in batteries. Batteries account for about 54% of cadmium in the solid waste stream. Fluorescent lights, Hg-vapor lamps, arc lamps, light switches, mirror coating, amalgam, and electrical apparatus are other sources of Hg in MSW, but to a much lesser degree than that of batteries (Bureau of Solid and Hazardous Waste, 1995; Tchobanoglous, 1993; Steinwachs, 1990). Table 2-4 identifies the discarding of Hg containing products into the MSW stream.

Waste Recovery and Resource Treatment

Combustion

Combustion is defined as the chemical reaction of oxygen with organic materials, to produce oxidized compounds accompanied by the emission of light and rapid generation of heat (Tchobanoglous, 1993). MSW combustion (incineration) has two main functions, (1) reduction in the volume of waste subject to final disposal, and (2) recovery of energy. Combustion facilities are referred to as waste-to-energy (WTE) units which are capable of producing steam and electricity

Table 2.4 Discard of Hg Containing Products in the MSW Stream (tons)

Product	United States		Florida	
	1989	2000	1989	2000
Household Batteries	621.10	98.50	32.30	5.12
Mercury Light Switches	0.40	1.90	0.02	0.10
Electric Lighting	26.70	40.90	1.39	2.13
Paint Residues	18.20	0.50	0.95	0.03
Fever Thermometers	16.30	16.90	0.85	0.87
Thermostats	11.20	10.30	0.58	0.54
Pigments	10.00	1.50	0.52	0.08
Dental Uses	4.00	2.30	0.21	0.12
Special Paper Coating	1.00	0.00	0.05	0.00

Adapted from Bureau of Solid and Hazardous Waste, 1995 and can be used in conjunction with source reduction, recycling and composting programs (USEPA, 1989). For MSW, excess air is used to ensure complete combustion of the organic fraction. This is represented by the equation

$$\text{Organic matter} + \text{excess air} \rightarrow N_2 + CO_2 + H_2O + O_2 + \text{ash} + \text{heat}$$

The endproducts of combustion include hot combustion gases and noncombustible residues. The heavy metals are normally found in the noncombustible residue known as ash. Heavy metals emissions from municipal waste combustion (MWC), particularly Hg, are cause for concern because significant amounts of Hg are released through incinerator stack emissions. Mercury can vaporize at very low temperatures (40-50°C), therefore it is easily emitted into the environment after which it can be converted into various forms, one of which is methylmercury which bioaccumulates in the food chain (Steinwachs, 1990). In 1986, EPA issued operational guidance on control technology for MWC. The final regulations were issued in December 1990 to reduce deleterious emissions. Batteries are supposed to be separated from MSW prior to incineration to reduce the heavy metal emissions. In the past, dead batteries in the waste stream were the principal source of heavy metals in the flue-gas stream (Bretz, 1990).

Metzger and Braun (1987) investigated in-situ Hg speciation in flue gas by liquid and solid sorption systems. Solid sorbents included iodated activated carbon and also gold or silver amalgams. Liquid sorbents included nitric acid/peroxydisulfate solutions. A condensation/absorption approach was used to identify Hg within the sorbents.

In Europe, Hg in gas emissions from waste incinerators consists of 20% of elemental Hg, 60% of divalent inorganic Hg compounds, and 20% of particulate Hg (Petersen et al., 1995). In Europe, the Hg content in MSW ranged from 0.3 to 9 g/t. In North America, the Hg content ranged from 0.36 to 5-8 g/t. Medical waste incinerators emit an average of about 30 g Hg/t in developed countries and 10-20 g Hg/t in developing countries (Pirrone et al., 1996).

Krishnan et al. (1997) investigated Hg control in MWCs and coal-fired utilities. The report showed the ability of activated carbons PC-100 and FGD to capture Hg^0 and $HgCl_2$ emitted from these two systems. The precursor for PC-100 is bituminous coal and for FGD, it is lignite. The PC-100 captured more Hg^0 than FGD at both temperatures for MWC and coal-fired simulations.

Carroll et al. (1995) investigated Hg emissions from a hazardous waste incinerator equipped with a State-of the-Art wet scrubber. Scrubber collection efficiency for Hg averaged 87% which was lower than expected. Gowin et al. (1993) studied the ability of triple-reverse-burned coal char (TRB char) to remove Hg vapor from the gasification reactor and determined that it was an effective sorbent for this purpose.

The main problem with MWC stems from the production of residual ash. The inorganic, noncombustible portion of the waste stream and the uncombusted organic matter are the constituents in the ash. Heavy metals remaining in incinerator ash are in a more leachable form. Exposure to these heavy metals occur in two ways. First of all, respirable particles may disperse into the air from the stack or during transport and handling. Next, leaching of the metals may occur at disposal, contaminating ground and surface water (Bretz, 1990). Fly ash and bottom ash often contain high levels of heavy metals that require special handling and burial in ash landfills. Standards for the handling, processing, disposal and recycling of MSW combustor ash in

Florida are found in Chapter 17-702, F.A.C., and were adopted by the Environmental Regulation Commission in June, 1990 (Department of Environmental Regulation, 1991).

Heavy metal concentrations and percentage distributions in soil samples confirm a strong tendency of heavy metals to accumulate in solid effluents and concentrate in fly-ash. This is one possible explanation of why there is a high amount of metals emitted per ton of burned waste (Morselli *et al.*, 1992). In the past, ash was just disposed of in a landfill specifically created for ash disposal called a monofill, but ash reuse is currently on the rise.

Ash recycling involves the use of ash in soil cement for road sub-base material, the use of ash as aggregate in road asphalt, and the construction of blocks from the ash for artificial reef construction (Department of Environmental Regulation, 1991).

Composting (Aerobic)

In MSW composting, preprocessing is performed to isolate the compostable portion, i.e., yard wastes, food wastes, and organic fractions such as paper. These materials constitute about 30 to 60 percent of the MSW stream. Separation of the compostable portion is generally performed using a rotating screen called a trommel. Once these are separated, they are usually shredded to reduce the particle size and moisture may be added to aid the composting process (USEPA, 1989). The main requirement for compost is that it should be suitable for agricultural use as an organic soil conditioner. Therefore, physical, chemical and biological stability, nonphytotoxicity and balance among mineral elements are the primary characteristics for compost to be useful to the soil and for crops (de Bertoldi et al., 1990).

Heavy metals do not degrade, but tend to be concentrated during the composting process. Metals of greatest importance are those which bioaccumulate, resulting in long or short-term toxic effects to organisms in the environment. Those most commonly regulated include Cd, Hg, Pb, Ni, and Zn. To date, most MSW composts have achieved regulatory limits for most metals, with the exception of lead, but there is an interest in attaining lower levels. The earlier that sorting occurs during the collection and composting process, the lower the heavy metal content in the finished compost. Source separation of a few contaminants such as lead acid batteries and television picture tubes will definitely decrease the heavy metal content of MSW compost (Richard and Woodbury 1994)

One report determining how composting affects heavy metal content stated that there was a great occurrence of water soluble heavy metals, except for Cd The concentrations of heavy metals were generally lower in the samples taken at the end of the composting period in the correctly prepared compost In correctly produced compost, maturation increases the humic acids with respect to the fulvic acids which does not occur in incorrectly produced compost This is an indication of the process of humification of organic matter leading to the unavailability of heavy metals as the heavy metals would now be complexed to the humic material, hence reaching the soil in a complexed, less mobile form Therefore, there would be a decrease in the leaching of heavy metals About one-third of the total content of heavy metals present in compost was reportedly bound to the alkali soluble organic matter (Canarutto et al 1991).

Heavy metal concentrations in the finished compost determine its usage Lower heavy metal content allows the compost to be used for practical applications, such as landscaping Higher metal concentrations in compost prohibit its usage because of the potential for plant uptake, or leaching into the environment (Steinwachs, 1990). Land application rate of heavy metal containing composts are dependent on the concentration of the metal, the pH of the soil and the cation exchange capacity of the soil. The decrease in concentration of metals in the soil compost mixtures results from leaching, with very little as a result of plant uptake.

In aerobic biologically stabilized solid waste residues one of the main forms of Hg will likely be mercuric oxide (HgO), along with other possible forms of Hg such as $HgCl_2$ which is likely to occur in the absence of sulfides. Chloride is normally a weak complexing agent, but it strongly complexes with Hg^{2+} and Hg_2^{2+} (Pohland et al., 1981). HgO may be dissolved and mobilized by acid rain percolating through residue-amended soil, increasing the chances of Hg being taken up by plants, as well as the possibility of leaching.

Landfilling

In anaerobic environments, such as sediments or landfills, or under mildly reducing conditions, there is a tendency for Hg to be precipitated as the sulfide (HgS). Mercuric sulfide is highly insoluble (log K_s = -50.02) (IUPAC, 1992), but can be transformed under low Eh and high pH conditions to the soluble form, HgS_2^{2-}, or to the free metal (Barkay, 1992; Wilken, 1992; Schuster 1991). The insolubility of HgS makes it resistant to methylation, but under aerobic conditions, HgS may be oxidized to the sulfate form which can then undergo methylation (Adriano, 1986).

Gases found in landfills include methane (CH_4), carbon dioxide (CO_2), ammonia (NH_3), carbon monoxide (CO), hydrogen sulfide (H_2S), nitrogen (N_2), and oxygen (O_2) (Tchobanoglous 1993). When considering the movement of Hg in the landfill and the transformations that occur, it can be hypothesized that dimethylmercury and elemental Hg may actually be emitted into the atmosphere. Anaerobic bacteria may act on the available Hg in the landfill and transform it to either monomethylmercury or dimethylmercury. The vapor pressure of monomethylmercury is quite low ($H < 10^{-4}$) while the vapor pressure of dimethylmercury is quite high ($H < 0.3$) (Barkay, 1992). As a result, this form of Hg may also be given off from a landfill in the same manner as methane or carbon dioxide which are the two most prevalent gases resulting from the anaerobic processes occurring in a landfill.

Under warm or hot conditions, Hg can be transformed to elemental Hg which is the second most volatile form of Hg (H=0.3), next to dimethylmercury. If this transformation does in fact occur in a landfill, then without proper control mechanisms, elemental Hg would be emitted into the atmosphere.

Heavy metals dissolved in aqueous systems exist in combination with other chemical species in the form of complexes. Metal ions, such as Hg^{2+}, combine with non-metallic compounds known as ligands by means of coordinate covalent bonds. In anaerobic residues, the species of Hg found will likely be mercuric sulfide (HgS). Cinnabar or mercuric sulfide is about the most stable Hg mineral that occurs in nature. This Hg mineral forms under reducing conditions where sulfate (SO_4^{2-}) has been reduced to sulfide (S^{2-}) (Mitra, 1986). Gambrell et al. (1978) showed that HgS is unavailable to plants. Therefore it may be feasible to use anaerobically stabilized solid waste residues for land application. If Hg is in the HgS form, there should be minor, if any, adverse impacts on crops, and the groundwater should remain uncontaminated as HgS is quite immobile and unlikely to leach.

The dumping of trash is becoming a less viable solution since the siting of landfills is extremely difficult as a result of public opposition The public invokes the NIMBY (not in my back yard) syndrome due to the odors and the potential contamination to surface and groundwater that may occur due to leaching However, as technology improves and liners become more reliable, landfills are meeting federal and state standards, which should actually ease the public's minds when it comes to the siting of landfills Some communities that are faced with diminishing landfill space send their waste to other states, a practice sanctioned by the Supreme Court ruling in 1978 (Philadelphia v New Jersey) that a state may not refuse waste from another state This poses a problem for those communities that are trying to extend the life of their landfills by recycling or reducing their own wastes when the space is filled by other counties or states (Concern, Inc , 1988)

The Solid Waste Management Act (SWMA), Senate Bill No 1192, was passed by the Florida Legislature and became effective on October 1, 1988 Under this Act, certain items were banned from landfills These are used motor oil which became effective October 1, 1988, lead acid batteries, which became effective on January 1, 1989, white goods which became effective on January 1, 1990, and yard trash which became effective January 1, 1992. Exclusion of these items from the landfill will reduce the amount of hazardous materials that can potentially contaminate groundwater or the surrounding land (Earle et al., 1991), especially heavy metals.

Landfill leachates often contain high concentrations of toxic heavy metals. Many of these metals can form strong complexes with biomolecules, therefore, even in small amounts their presence can have adverse effects on both plants and animals (Bolton and Evans, 1991). Some probable ligands in landfill leachates and residues would be chlorides, sulfates, phosphates, and organics (Pohland et al., 1981). Table 2-5 shows the median concentrations in MSW leachate, in comparison with existing exposure standards. It was interesting to note that on an overall basis, the USEPA (1988) report had lower levels than those given in the Congress of the United States (1989) report. The fact that certain items containing heavy metals were banned in 1986 and 1989 would have led one to believe that the heavy metal contents would have decreased. Obviously, items such as batteries are still finding their way into landfills even though there is now an emphasis on rerouting them away from landfills and recycling the relevant

Table 2.5 The median concentrations in MSW leachate, in comparison with existing exposure standards

Metals	Median concentration (ppm)	Exposure standards (ppm)
Antimony	0.066	0.01
Arsenic	0.042	0.05
Barium	0.853	1.0
Beryllium	0.006	0.2
Cadmium	0.022	0.01
Chromium	0.175	0.05
Copper	0.168	0.012
Iron	221	1000
Lead	0.162	0.05
Manganese	9.59	0.05
Mercury	0.002	0.002
Nickel	0.326	0.07
Selenium	0.012	0.01
Silver	0.021	0.05
Thallium	0.175	0.04
Zinc	8.32	0.110

(USEPA, 1988 and Congress of the United States, 1989) metals. Landfill leachates contain many organic and inorganic ligands, particularly chloride (Cl^-), which help to determine the forms of metals in solution. High concentrations of inorganic ligands, primarily chloride (Cl^-), and high dissolved organic carbon (DOC) concentrations affect the speciation of metals in landfill leachates. Complexation reactions in solution lower the positive charge associated with the metal and therefore may affect the mobility of leachate metals in the soil and underlying sediments (Bolton and Evans, 1991). Therefore, methods have to be developed to decrease the amount of heavy metals in leachate.

Leachate recycling is one method to decrease the amount of heavy metals present. One report showed that leachate recycling did not result in an increase in heavy metal concentrations, but actually removed some of the heavy metals in the solid waste columns. The explanation provided was that under anaerobic conditions that exist in a landfill, heavy metals precipitate as sulfides (Pohland *et al.*, 1979).

Anaerobic Digestion

The main steps in anaerobic digestion of MSW are: (1) Pretreatment - to remove undesirable material, upgrade and homogenize the feedstock for digestion and to protect downstream treatment processes. (2) Anaerobic digestion - to produce biogas for energy and to deodorize, stabilize, and disinfect the digestate product. (3) Post-treatment - to complete the stabilization and disinfection of the digestate, to remove residual inert undesirable material (glass and plastics) and produce a refined product of suitable moisture content, particle size and physical structure for the proposed end-use. The end uses are energy in the form of biogas, typically around 100-200 m^3 biogas per ton of organic MSW digested, also solid and liquid by-products which can be used as compost products and have a value as a fertilizer or soil improver (IEA Bioenergy, 1994).

The majority of the literature on anaerobic digestion deals with zinc (Zn^{2+}), iron (Fe^{3+}), cadmium (Cd^{2+}), copper (Cu^{2+}), chromium (Cr^{3+}, Cr^{6+}) Nickel (Ni^{2+}), and lead (Pb^{2+}). The focus of most of these articles was the inhibition of anaerobic digestion by these heavy metals. It was determined that concentrations and species of these metals were indicative of their toxicity and the ability of microorganisms (bacteria) to adjust to their effects. Temperature, pH, hydraulic retention time, and the ratio of the toxic substance concentration to the bacterial mass concentration are determinants is the inhibitory concentrations of these heavy metals. Some inhibitory concentrations given were Cd^{2} (180 ppm), Fe^{3+} (1750 ppm), Cu^{2+} (170) ppm, Cr^{6+} (450 ppm), Cr^{3+} (530 ppm), and Ni^{3+} (250 ppm) (Chynoweth and Pullammanappallil 1996, IEA Bioenergy, 1994; Peiffer, 1993, Lin, 1993, Lin 1992; Mueller and Steiner, 1992).

Determination of the heavy metal content after the digestion process is the principal determining factor in the feasibility of using the liquid or solid digestate as compost. Post-treatment follows the actual anaerobic digestion process which allows for two to four weeks of maturation for the solid fraction whereas the liquid fraction may be directly applied onto farmland as slurry, if it is of good quality and meets all set regulations. Review of the literature provided no information about the presence of Hg. Hence, it is obvious that Hg has rarely been included in past studies.

In an anaerobic environment, microorganisms that do not need oxygen are called anaerobes. There are two types of anaerobes, obligate and facultative. Obligate anaerobes cannot use oxygen at all, and tend to be poisoned by it. Facultative anaerobes are able to use oxygen when it is present, but can use other sources when oxygen is absent. The other sources that replace oxygen are nitrate (NO_3^-), sulfate (SO_4^{2-}), and carbon dioxide (CO_2). When these compounds are used in the energy-generating process they are reduced. Nitrate is reduced to nitrogen gas (N_2), sulfate is reduced to hydrogen sulfide (H_2S), and carbon dioxide is reduced to methane (CH_4). Bacteria that use nitrate are called denitrifying bacteria and those that use sulfate are called sulfate reducers, and those that use carbon dioxide are called methanogenic bacteria (methanogens). Denitrifying bacteria are facultative anaerobes and sulfate reducers and methanogenic bacteria are called obligate anaerobes (Brock and Madigan, 1988; Brock and Brock, 1978).

In an MSW anaerobic digester, there is a consortium of microorganisms that play a part in methane fermentation. Large-molecular weight compounds such as polysaccharides, proteins, and fats, are converted to methane by these microorganisms. For conversion of a typical polysaccharide, this consortium includes cellulolytic bacteria which cleave high molecular weight cellulose molecule into cellobiose and free glucose, fermentative anaerobes which ferment the glucose to a variety of products including acetate, propionate, butyrate, hydrogen (H_2), and CO_2. Methanogenic bacteria consume any H_2 that is produced. Also, certain methanogens are capable of converting acetate to methane (Brock and Madigan, 1988).

Other organisms that convert complex materials to methane are H_2-producing fatty acid-oxidizing bacteria which use fatty acids or alcohols as energy sources and grow very well in the presence of H_2-consumers such as methanogens and sulfate reducers. The association between hydrogen producers and hydrogen consumers is known as interspecies hydrogen transfer which is important in the anaerobic digestion process. *Syntrophomonas* and *Syntrophobacter* are H_2-producers which generate acetate, CO_2, and H_2. Hydrogen-consuming acetogenic bacteria (*Acetobacterium*) consume hydrogen and produce acetate for methanogenesis. During methanogenesis, methanogens reduce the acetate, CO_2 and H_2 to methane and carbon dioxide which are the main end products of anaerobic digestion (Chynoweth and Pullammanappallil, 1996, Brock and Madigan, 1988).

This literature review has included all three phases of the environment, namely air, earth, and water, and the many Hg interactions that take place in the environment. This literature review showed that data of Hg in landfills and MSW is extremely limited. However, data on metal concentrations in landfill leachate is well documented. The fact that Hg-containing devices are still finding their way into landfills in spite of more stringent recycling efforts, is the primary reason why more information is needed on the fate of Hg in landfills. The literature on anaerobic degradation processes strongly suggests that transformation of inorganic and elemental Hg to volatile organic forms is highly likely to occur in landfills.

CHAPTER 3
MATERIALS AND METHODS

Phase I Hg in the Alachua County Landfill

Samples that were analyzed for total Hg in this study had been obtained from the Alachua County Landfill during a previous study (Miller *et al.*, 1996; Miller *et al.*, 1994). Five areas were sampled and were designated as LFS1, LFS2, LFS3, LFS4, and LFS5 (Figure 3-1). LFS1 is a leachate recycle area (Infiltration Pond 1), LFS2 is a control area for leachate recycling, LFS3 is a 4.5 hectares closed landfill, LFS4 is a 12 hectare closed landfill, and LFS5 is a leachate recycle area with horizontal injection. The active lined landfill (11 hectares) at this site began receiving MSW in 1988. Leachate recirculation was performed on a portion of this lined landfill (LFS1) which is adjacent to the leachate recycle area. LFS2 is actually a portion of the lined landfill that was used as a control area where there was no

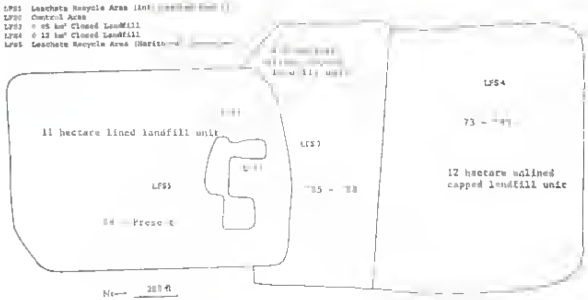
LFS1 Leachate Recycle Area (Unl
LFS2 Control Area
LFS3 0.05 km² Closed Landfill
LFS4 0.12 km² Closed Landfill
LFS5 Leachate Recycle Area (Marit
lined landfill unit
11 hectare lined landfill unit
LFS5
Present
LFS3
'85 - '88
LFS4
73 -
12 hectare unlined capped landfill unit
N
200 ft

Figure 3.1 Alachua County Landfill (Lee, 1996)

leachate recirculation occurring LFS3 received waste from 1985 to 1988 and LFS4 received waste from 1973 to 1985 and both were in a portion of the landfill that was unlined

Solid waste samples were collected from LFS 1 both prior to and following the filling of the infiltration pond with leachate Three boreholes were normally augured at each site with a 10 16 cm truck mounted open flight power auger that penetrated through the landfill until it reached the desired depth In general, four samples were collected per hole at 3 05 m (10 feet) intervals The maximum depth of the boreholes ranged from 9 14 - 13 72 m The top portions of some samples (0 - 1 52 m) were discarded because they were mostly cover soil Table 4-1 shows the depths of each collected sample Additional information on sampling procedures can be found in Miller et al (1996)

After collection, the samples were placed into polyethylene bags and brought back to the Bioprocess Engineering Research Laboratory of the Agricultural and Biological Engineering Department at The University of Florida and stored in freezers until processed Processing of these samples involved separation and characterization of waste components, moisture content, and solids composition determinations and biochemical methane potential assays. The samples were dried at 105°C, grossly picked, and then passed through a 6 mm screen. The material that remained on the 6 mm screen was finely picked and ground to 0.76 mm using an Urschell Mill (Comitrol Model 3600). The material that passed the 6 mm screen was then passed through a #40 screen (0.42 mm). The material that remained on the #40 screen was finely picked and then ground to 0.76 mm using the Urschell Mill. For the purpose of total Hg analyses, the samples consisted of three fractions. One fraction was that remaining on the 6 mm screen, a second fraction was that passing the #40 screen, and the third fraction was that remaining on the #40 screen. These are shown in Table 4-1. These three fractions represented the entire sample taken from the landfill after gross picking and fine picking. In the study by Miller et al. (1996) the SW from the landfill was also characterized as paper, cardboard, food waste, glass and stone, metal, wood, plastics and rubber, and fabrics. Gross and fine picking removed glass and stones, metal, wood, plastics, rubber, and fabrics, leaving essentially paper, cardboard, and food wastes.

The elaborate separation scheme used by Miller et al (1996) was developed to obtain the fraction of SW that is directly responsible for methane production namely the volatile organic fraction This is shown in Figure 3-2 For Phase I of this study, the three fractions described above were analyzed to evaluate total Hg concentrations existing in the landfill and to give an idea of the Hg distribution that occurred in such an environment Mercury was being evaluated because there has been little research concerning Hg in biologically stabilized SW residues The fact that these samples are volatile organic fractions are important concerning Hg because Hg binds strongly to organic material, therefore the majority of the Hg in the landfill should be bound to any existing organic matter present

Total Hg in these samples was measured on a ng/g dry weight basis to give an idea of the Hg levels existing in the Alachua County Landfill These Hg concentrations could have been converted back to a wet weight basis, but it was not deemed necessary since it is not scientifically incorrect to report on a dry weight basis Furthermore, if the landfill is ever reclaimed and the SW applied to land, application rates will be computed on a dry weight basis It must also be noted

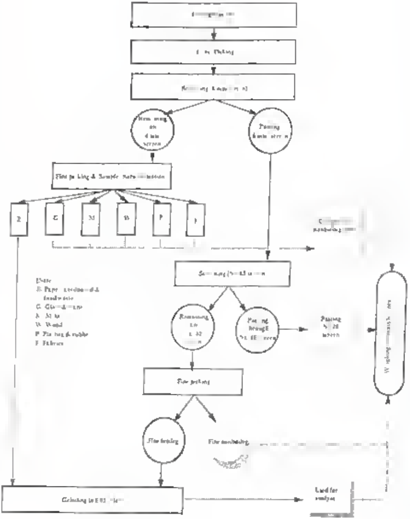
G
M
W
P
W Wood
F Fabrics
Fine picking
Used for analysis

Figure 3-2 Sample Handling Procedure (Lee 1996)

that the Hg values reported in this study may be lower than the actual values because the samples had been dried at 105°C in the study of Miller et al. (1996). Elemental Hg vaporizes at around 40-50°C and any elemental Hg that may have been in the original landfill samples would have been lost in the drying process.

Total Hg was determined using standards ranging from 0 to 550 ng Hg. A check sample was used in every run as a way to evaluate whether or not the system was performing adequately and also as a reference point to the validity of the other samples. A duplicate and a spike were run with each sample batch (12 samples) and a continuous calibration blank and a continuous calibration variance (a 50 ng Hg spike of a blank sample) were used throughout every sample batch to ensure that the instrument absorbances were being maintained. The acceptance criteria for the calibration curves (6 standards, 1 blank) was $r^2 > 0.99$. The blanks had to be below the Method Detection Limit, the spikes had to be +/- 20% of the true value, and for duplicate samples, the standard error had to be < 20%. This QA/QC was used in a previous Hg study (Delfino et al., 1993). Results of the QA/QC are given in Appendix A.

Total Hg in the SW fractions was determined using the digestion procedure described in EPA Method 7471 for the determination of Hg in soil and sediment. Following the degestion procedure, Hg was analyzed by cold vapor atomic absorption spectrophotometry (USEPA, 1986). Two grams of sample were weighed to the nearest 0.0001 g on an enclosed Mettler analytical balance. The sample was transferred quantitatively to an acid rinsed 300 mL BOD bottle with a 10 mL double distilled deionized (DDDI) water rinse. The digestion involved 2.5 mL of trace metal grade concentrated nitric acid and 5 mL of trace metal grade concentrated sulfuric acid. The sample was heated at 95°C for two minutes, then 15 mL of potassium permanganate (50 g/L), and 8 mL of ammonium peroxydisulfate (50 g/L) were added to the digestion mixture. The sample was heated at 95°C for one to two hours. An additional 15 mL of potassium permanganate was added if the color disappeared within fifteen minutes of the initial addition. Upon completion of digestion, samples were cooled and decolorized by the addition of 6 mL of hydroxylamine hydrochloride solution (120 g hydroxylamine sulfate and 120 g sodium chloride per liter of deionized water solution). Figure 3-3 shows a schematic of the procedure.

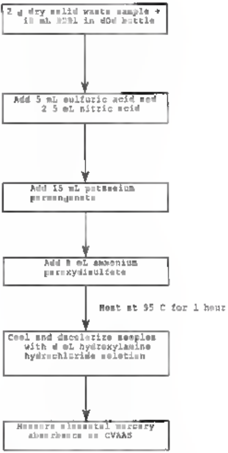
2 g dry solid waste sample + 10 mL DDW in BOD bottle
Add 5 mL sulfuric acid and 2.5 mL nitric acid
Add 15 mL potassium permanganate
Add 8 mL ammonium peroxydisulfate
Heat at 95 C for 1 hour
Cool and decolorize samples with 6 mL hydroxylamine hydrochloride solution
Measure elemental mercury absorbance on CVAAS

Figure 3-3 Total mercury analysis of solid waste fractions as described in EPA Method 7471

Trace metal grade nitric and sulfuric acid are oxidizers Potassium permanganate is another strong oxidizer that was added to eliminate possible interference from sulfide Ammonium peroxydisulfate is another added oxidizer to ensure the complete oxidation of the sample Heating the sample in a water bath, along with the strong oxidizers, converts all of the Hg to Hg^{2+}

Each digested SW sample was transferred to a plastic reaction vessel fitted for a Perkin Elmer MHS-10 cold vapor unit The sample was purged with high purity nitrogen gas Stannous chloride was used to reduce Hg^{2+} to the elemental state, Hg^{0} Elemental Hg was then swept into an open ended quartz tube (1 cm diameter) with a 16 cm cell path length The Hg was quantified by CVAAS using a Perkin Elmer Model 3030 Atomic Absorption Spectrophotometer (λ = 253 6 nm, SBW = 0 7 nm) with Hg hollow cathode lamp (I = 6 MA) The standard calibration curve working range (0 to 550 ng) gave an absorbance range of 0 017 to 0 260

Total Hg (ng/g) was determined using the equation derived from the regression analysis of the Hg standard curve From the equation

$$y = mx + b$$

Mercury (ng) was obtained using the noted absorbance from the atomic absorption minus the y-intercept divided by the x-coefficient i.e.,

$$(abs - b)/m$$

Since the SW samples from the Alachua County landfill had been dried prior to Hg analysis then the determination of Hg(ng/g) involved Hg(ng) divided by dried weight (g) measured out prior to analysis.

The Hg stock solution (1000 ppb) was made using 0 1 mL of 1000 ppm ±1% Mercuric Nitrate where 1 mL = 1 mg Hg (Fisher Scientific) and adjusting the volume to 100 mL in a 100 mL volumetric flask. The top of the volumetric flask was sealed with parafilm and the entire flask was enclosed in aluminum foil to slow any possible degradation that may occur due to the infiltration of light

Successive dilutions of the stock Hg solution were made to obtain a working standard containing 10 ppb or 100 ppb This working standard was prepared fresh daily prior to sample preparations. Acidity of the working standard was maintained at 0 15% trace metal grade nitric acid.

Aliquots were taken from the working standard to develop a range of standards that the sample data should fall within. The Hg standards were adjusted to a total of 10 mL with DDDI water. Next, the same procedure as stated above was repeated, i.e., the addition of the acids, potassium permanganate, ammonium peroxydisulfate, the digestion in the water bath at 95°C, and finally the addition of sodium chloride hydroxylamine sulfate. The stannous chloride on the MHS-10 unit was used as the reducing agent and the elemental Hg absorbances were then noted on the AAS. Due to the toxicity of Hg vapor, it was imperative that there be a well working vent over the instrument during analyses.

Compost samples were obtained from Dr. D. Graetz in the Soil and Water Science department at the University of Florida. These were previously collected from the Palm Beach County compost facility and consisted of a 1:1 mixture of biosolids to yard waste. These samples were air-dried, ground, and total Hg analyses were performed using EPA Method 7471. The Hg concentrations in these aerobic compost samples were compared with those found in the SW from the Alachua County Landfill. The raw data for the Palm Beach County compost samples are provided in Appendix A.

Phase II -Landfill Simulated Anaerobic Reactors

Reactor design

A schematic of the reactor design is shown in Figure 3.4. Each reactor was made from PVC pipe measuring 50.8 mm diameter and 635 mm long. PVC threaded end caps with inserted plastic threaded tees were used at either end of the reactor. There was a 38.1 mm diameter and 304.8 mm long schedule 40 PVC insert which contained the SW. The insert had holes drilled throughout it to enable the free movement of leachate into and out of the SW. At the bottom end of the insert there was a glass fiber mesh to hold the SW intact, but at the top it was left open. Two additional 38.1 mm diameter PVC inserts were placed at either end of the SW insert to decrease the amount of SW movement in the leachate circulation process. The additional inserts also had glass fiber mesh at the ends to keep the SW in place. The plastic tee at the bottom of the reactor had septa that were secured with copper wire and/or clamps to allow for the collection of leachate. The tee at the top of the reactor had a septum at one end for the collection of gas samples via syringe. The other end of the tee had a short piece of 6.35 mm diameter flexible tygon tubing (Norton Co.) to which a 3-way valve was inserted,

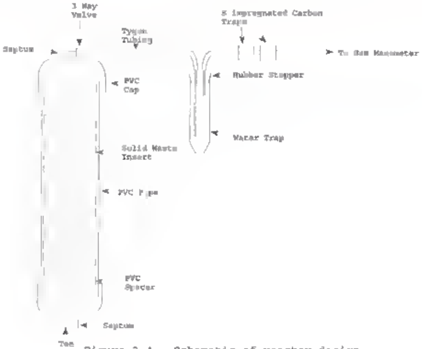
3 Way Valve
Tygon Tubing
Septum
PVC Cap
Solid Waste Insert
PVC Pipe
PVC Spacer
Septum
Tee
5 Impregnated Carbon Traps
To Gas Manometer
Rubber Stopper
Water Trap

Figure 3-4 Schematic of reactor design

followed by tygon tubing that led into a large test tube fitted with a 2-hole rubber stopper that served as a water trap to prevent any leachate from entering the carbon traps. Tygon tubing then connected the water trap to two sulfur impregnated carbon traps in series. The sulfur impregnated carbon (10 12% Sulfur) in the traps was obtained from Norit Americas and the grade was Norit RBHG 3 and the sample number was A9805. The encasing for the traps was predrying tubes with end caps (Bel Art Products). The tubing from the carbon traps then went into an inverted 1000 mL plastic graduated cylinder fitted with a 2-hole rubber stopper which served as a gas manometer. The gas manometers contained an aqueous 5% hydrogen chloride (HCl) solution for the purpose of reducing the amount of CO_2 being dissolved.

The acid solution in the gas manometers which was displaced by gas generated in the reactors was connected to a main acid solution reservoir consisting of a 1000 mL graduated cylinder, which was open to the atmosphere. This main reservoir served to refill the gas manometers with acid solution as needed. To measure the amount of gas produced by each reactor, the main reservoir was moved over to each inverted graduated cylinder and their liquid levels were matched and then noted, ensuring that all gas volumes were measured at atmospheric pressure The reactors were incubated at 50 ± 2°C Figure 3-5 shows the entire setup of the system Figure 3-6 shows a view of the water traps that prevented leachate from entering the carbon traps

Figure 3-7 shows how the solid waste insert was placed into the encasing Figure 3-8 shows the glass fiber mesh at the bottom of the SW inserts Figure 3-9 shows the top part of the system including the carbon traps Figure 3 10 shows a close-up view of the carbon traps There were two reactor runs performed using this system

Solid Waste

The SW used in this study was artificially made to represent the acutal landfill SW left behind when metals, glass, plastics and wood have been removed i e , grass and fine picked. The artificial waste consisted of office paper, newspaper, and dog food, the last ingredient being added to represent the food portion in an MSW sample The office paper and newspaper were finely shredded The dog food consisted of dry pellets that were first mixed with the shredded paper ground, and finally the entire mixture homogenized as well as possible

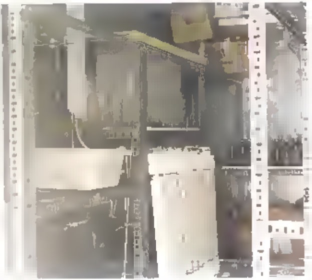

Figure 3 5 Entire setup of the reactor system

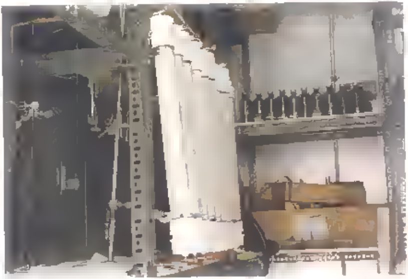

Figure 3 6 Liquid collection traps behind the reactors

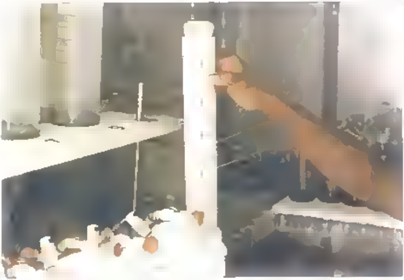

Figure 3.7 Solid Waste Insert [illegible]

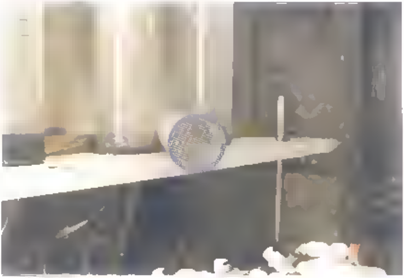

Figure 3.8 [illegible] mesh at bottom of solid waste insert

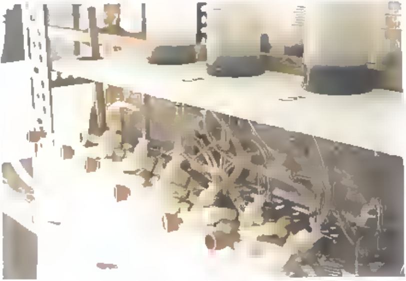

Figure 3-9 Top view of the reactor system

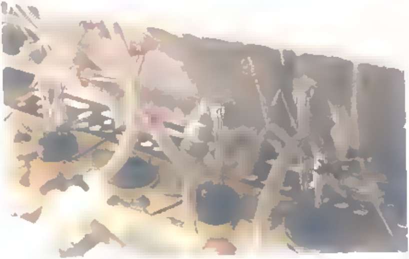

Figure 3-10 Close-up view of the activated carbon traps

The insert was packed with 60 g of the artificial SW at a packing density of 300 lb/yd^3. This density was chosen because it represents the density of SW in a packing truck, i.e. 300-400 lb/yd^3. The packing involved determining the volume of the insert and how much SW would be needed to yield a packing density of 300 lb/yd^3. Volatile solids analyses on the SW were performed in triplicate according to *Standard Methods for the Examination of Water and Wastewater, 1989, Section 2540E* Fixed and Volatile Solids, pp. 2-77.

Reactor operation

The nine reactors were all operated in the same manner. During setup, the SW was spiked with Hg prior to being placed into the reactor inserts. Spiking was done by adding the appropriate volume of a 100 µg Hg/L standard to the SW. These were then left to sit for about an hour at room temperature so that the Hg could bind to the SW. Next, these inserts were placed into the reactors and 600 mL of leachate from another digester was used as the innoculum in each reactor. Initial analyses of methane, pH, VPAs, volatile solids and Hg levels were perfomed on separate samples of the SW and the leachate.

In the first run, the first three reactors were used as controls. They contained the SW and the leachate, but they were not spiked with Hg Three replicates were used at two different Hg levels, namely 100 ng Hg and 2000 ng Hg. In the second run the first three reactors were once again used as controls, but two replicates were used at three different Hg levels, namely 100, 1000 and 2000 ng Hg In both runs, a total of nine reactors were used At the same time on a daily basis the liquid levels in the gas manometers were noted to determine the amount of gas produced by the reactors in a 24 hour period. The valves leading from the reactors were then closed off to the reactors to allow for the collection of gas samples without introducing air to the system After gas sample collection, the reactors were inverted to allow for recirculation of the leachate and also to flush out the volatile fatty acids. The reactors remained inverted for a period of about 30 minutes and then returned to their original position after which leachate samples were collected Liquid levels in the gas manometers were then reset for the next day's readings

The carbon traps were removed at the end of the reactor run for the analysis of Hg in the gas phase The leachate was also analyzed for Hg at the completion of the reactor run It was imperative that the solids not be sampled until the run was complete, otherwise the entire system would be disturbed as air would be introduced into the system. The septa at the top and the bottom tees of the reactors were changed every couple of days as the constant sampling would create holes large enough for the introduction of air into the system, thus causing destabilization of the redox conditions. General maintenance was performed on a regular basis, such as tightening the ties on the tygon tubing, tightening the screw caps on the reactors, and checking the gas manometers for liquid leaks. The temperature on the incubator was also checked regularly to ensure that it was being maintained at 50°C. All readings and sampling were taken inside the incubator at this temperature. The only time that the incubator door was open was when a person was entering or leaving. It was never left open for any amount of time so that the temperature could be maintained.

MSW Analysis

Total solids and volatile solids analyses were conducted in triplicate. A portion of the sample was dried at 105°C for 24 hours to determine the moisture content, after which it was placed in a 550°C oven to ash. The difference between the dried solids and the ashed solids provided volatile solids present in the sample. A smaller percentage of volatile solids in a sample would indicate less organic matter and hence, a more highly stabilized sample. Volatile solids indicates the potential degree of waste decomposition.

Gas Sampling and Analysis

Gas samples were taken at the same time each day for the first seven days, and then every other day afterwards. The samples were taken prior to the recycling of leachate. The septum at the top tee of the reactor allowed for access with a 10 mL syringe. Samples were taken to the laboratory right after collection and analyzed immediately. These samples were run in triplicate for statistical purposes.

Gas composition analysis for CH_4, CO_2, O_2 and N_2 was measured on a Fisher Model 1200 Gas Partitioner chromatograph equipped with two stainless steel columns and a thermal conductivity detector. A 10 mL aliquot of gas was injected manually into a sampling loop which conducted sample into each of two columns. The first column was 2 m x 3.2 mm packed with 80/100 mesh Poropak Q (Supelco Inc.) used for the separation and detection of CO_2, and the second column was 3.35 m x 4.8 mm packed with 60/80 mesh molecular sieve 13X (Supelco, Inc.) for the separation of O_2, N_2, and CH_4. The column temperature was 50°C, the detector bridge current was 175 mA, and the attenuator was set at 4. Helium was the carrier gas at a flow rate of 30 cm^3/min. Standard gas samples with known concentrations of the four compounds were initially injected to make sure that the instrument was performing properly.

Volatile Fatty Acids

Standards Preparation

To a tared 10 mL volumetric flask, 1 g isobutyric acid was added and the weight recorded. The flask was filled to the line with DDDI water and mixed well. To a 1 L volumetric flask, 200 mL DDDI water was added. One hundred mL of Phosphoric acid was added slowly to the water in the 1 L flask. The isobutyric acid solution was then added to the 1 L flask and the 10 mL flask was rinsed six times with DDDI water and the rinsate was added to the 1 L flask. The 1 L flask was then filled to the line with DDDI water. This isobutyric acid solution was decanted into a 1 L amber bottle and sealed with a teflon cap. This bottle was then labeled "VFA Internal Standard" and dated.

Next, another 10 mL volumetric flask was tared and 1 g each of acetic, propionic, butyric, isovaleric, and valeric acids were added to the flask. Each weight was recorded and rezeroed between acids. The 10 mL flask was then filled to the line with DDDI water and mixed. To a 1 L volumetric flask, 500 ml of DDDI was added. Once again, the 10 mL flask was rinsed six times with DDDI and the rinsate added to the 1 L flask. The 1 L flask was then filled with DDDI water and mixed well. The acids solution was then decanted into a 1 L amber bottle and sealed with a teflon cap. It was labelled "VFA Calibration Standard" and dated. The VFA internal standard solution and the VFA calibration standard solution were kept refrigerated between uses. These were made fresh prior to the start of the first run and were kept in a refrigerator for preservation purposes.

Leachate Sampling

Leachate samples were collected for the first seven days of the reactor run and then every other day after that. Samples were collected through the septum at the bottom tee of the reactor after the recycling of leachate. This sampling was performed using a needle on a 10 mL syringe. Nine mL of the unfiltered leachate sample was stabilized with 1 mL of 85% H_3PO_4. This was then distributed into three 2 mL centrifuge tubes and spun down in a centrifuge at 3000 RPM for at least three minutes. To an 8 mm gas chromatograph glass sample vial, 111.1μL of the internal standard, isobutyric acid, was added, followed by 1 mL of the centrifuged supernatant sample. The sample vials were then sealed and mixed by vortex. At this point, the sample was ready for analysis. These samples were run in triplicate.

Leachate Analysis

A Shimadzu gas chromatograph/flame ionization detector (GC/FID) fitted with an auto sampler and equipped with a 1.8 m x 2 mm glass column of 10% SP-1000 on 100/120 Chromosorb W/AW (Supelco, Inc.) was used for VFA analysis (Chynoweth *et al.*, 1990). The column was maintained at 180°C, the injector at 180°C, and the flame ionization detector at 200°C. Nitrogen gas was used as the carrier gas at a flow rate of 30 cm^3/min. Air was used to operate the autosampler and also the FID. A Perkin Elmer Model integrator was used to calculate the concentration in mg/L of the VFA's. The VFA's that were analyzed were acetate, propionate, isobutyrate, butyrate, isovalerate, and valerate. One microliter of sample was injected into this GC.

pH Analysis

Prior to VFA analysis, pH was determined on the leachate samples that were collected. A Fisher pH meter, model #805 MP was used after calibration with pH 4.0 and 9.0 buffers.

Mercury Analyses

MSW Mercury Analysis

Total Hg analysis was performed on the SW prior to and after the reactor run, thus providing initial and final Hg concentrations in the SW. This involved using the digestion procedure described in EPA Method 7471.

The solid samples were collected at the end of the reactor run. The SW was placed in a ziploc freezer bag and mixed as well as possible. If analysis was not going to be performed at that time, then the bags were placed in the freezer until then. The normal holding time for these samples was 28 days.

Carbon Trap Sampling

The carbon traps were removed by clamping both ends of the tygon tubing entering and leaving the traps so as not to allow for the introduction of air. When the traps were removed, the ends were sealed with parafilm and taken to the lab for the carbon to be analysed by CVAAS. Activated carbon (especially sulfur and iodine impregnated activated carbon) has been distinguished as an excellent sorbent for elemental Hg (Vidic and McLaughlin 1996, Krishnan et al , 1994, Moffitt and Kupel, 1971).

Carbon Trap Analysis and Method Development

A method had to be developed for the determination of Hg in the carbon traps The carbon in the traps was impregnated with sulfur which binds Hg tightly to produce one of the most stable forms of Hg, mercury sulfide, when the Hg in the gas phase was passed through it The usual EPA method for the determination of Hg in solid material had to be tested and modified as necessary to achieve full recovery of Hg from the carbon traps

Prior to running the reactors, the traps were built and tested for their ability to trap elemental Hg This was performed by reducing a known Hg standard in solution to the elemental form by using the reductant stannous chloride in the MHS-10 cold vapor unit. The standard samples were analysed to note the absorbance readings and then the sulfur impregnated carbon traps were put on the line leading from the cold vapor unit to the quartz cell and the absorbances were noted again It was determined that the traps did in fact trap all of the elemental Hg being passed though the tube since trapping completely eliminated any absorbance as measured by the AA

It was then observed that the EPA digestion procedure described in Phase 1 could not recover any of the Hg caught on the carbon traps The literature was exhausted for means to remove Hg from the S impregnated carbon traps, but no reports were found. It apparently required a stronger digestion step than the EPA Method 7471 used. In the original method, 10 mL of DDDI water. 2 5 mL of nitric acid, 5 mL of sulfuric acid. 15 mL of potassium permanganate and 8 mL of ammonium peroxydisulfate were used in the Hg digestion of the carbon traps. The first modification of the method involved using 10 mL of distilled deionized water, 5 mL of nitric acid, 10 mL of sulfuric acid. 30 mL of potassium permanganate and 16 mL of ammonium peroxydisulfate, but no Hg was again recovered from the carbon traps The final modification of the method used 10 mL of DDDI water, 7 5 mL of nitric acid. 15 mL of sulfuric acid, 45 mL of potassium permanganate, and 24 mL of ammonium peroxydisulfate and the time in the water bath lengthened to over two hours This time, the Hg recovered from the traps was >98% The modified digestion procedure was repeated to confirm these recoveries and they were found to be reprocible.

Figure 3 11 shows a schematic of the method for the analysis of Hg in sulfur impregnated carbon. The data for the method development for Hg analysis of the carbon traps are presented in the Results section

Mercury in Leachate

All of the leachate at the end of the reactor runs was collected by removing one of the bottom septums and letting it flow into an acid rinsed bottle which was then taken to the lab for analyses If analyses were not going do be done right away, the samples were placed in a freezer until such time

The Hg in the leachate samples was determined using EPA Method 7470 which is a cold vapor atomic absorption procedure that is used for the determination of Hg in mobility-procedure extracts, aqueous wastes, and groundwaters. It can also be used for the analyses of solid sludge-type wastes, even though EPA Method 7471 is better suited for sludge analyses.

EPA Method 7470 involved transferring 100 mL of the sample, in this case, leachate, to an acid rinsed 300 mL BOD bottle Added to this leachate sample were 5 mL of concentrated sulfuric acid and 2 5 mL of concentrated nitric acid The sample was heated at 95°C for two minutes, then 15 mL of potassium permanganate (50 g/L), and 8 mL of ammonium

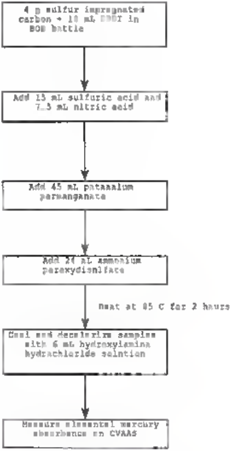
4 g sulfur impregnated carbon + 10 mL DDDI in BOD bottle
Add 15 mL sulfuric acid and 7.5 mL nitric acid
Add 45 mL potassium permanganate
Add 24 mL ammonium peroxydisulfate
Heat at 95 C for 2 hours
Cool and decolorize samples with 6 mL hydroxylamine hydrochloride solution
Measure elemental mercury absorbance on CVAAS

Figure 3-11 Modification of EPA Method 7471 for determination of total mercury in sulfur-impregnated carbon.

peroxydisulfate (50 g/L) were added to the digestion mixture The sample was heated at 55°C for one to two hours It was not necessary to add any extra potassium permanganate as the leachate samples were not prone to change color Upon completion of digestion, samples were cooled and decolorized by the addition of 6 mL of hydroxylamine hydrochloride solution (120 g hydroxylamine sulfate, and 120 g sodium chloride per liter of deionized water solution) Figure 3-12 shows a schematic of the procedure

Tygon Tubing Experiment for Mercury

Another experiment tested the ability of tygon tubing at room temperature (24C) to absorb elemental Hg since tygon tubing was used to connect the carbon traps to the anaerobic digesters This experiment was performed to determine the role that tygon tubing may or may not play in the entrapment of elemental Hg Tygon tubing was attached to the cold vapor unit leading to the quartz cell and the absorbances of known Hg standards were noted on the CVAAS The tubing did not appear to have any effect on the Hg absorbances, thus it did not appear to absorb elemental Hg.

An additional experiment was performed to determine whether tygon tubing could trap elemental Hg and Hg^{2} at 50°C

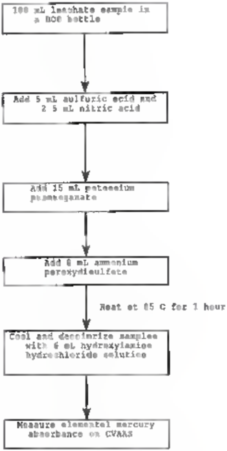
100 mL leachate sample in a BOD bottle
Add 5 mL sulfuric acid and 2.5 mL nitric acid
Add 15 mL potassium permanganate
Add 8 mL ammonium peroxydisulfate
Heat at 95 C for 1 hour
Cool and decolorize samples with 6 mL hydroxylamine hydrochloride solution
Measure elemental mercury absorbance on CVAAS

Figure 3-12 Total mercury analysis in leachate samples as described in EPA Method 7470

and whether Hg^{2+} could volatilize at 50°C and be trapped on the S-impregnated activated carbon. This was a 60 hour experiment that was set up in an incubator room that operated at 50°C. There were two pairs of flasks each connected in series to a pair of S-impregnated activated carbon traps. An aquarium pump with a total flow rate of 30 ml/min was attached to the two pairs of flasks. All four flasks contained DDDI water and were spiked with 100 ng Hg^{2+} as $Hg(NO_3)_2$. To one pair of flasks, $SnCl_2$ was added to reduce the Hg^{2+} to Hg^0. No $SnCl_2$ was added to the other pair, leaving the Hg in the +2 valence form.

Although these experiments showed that tygon tubing did not trap either elemental or divalent Hg, the tygon tubing that ran between each reactor and the carbon traps was weighed, cut up into small pieces and digested as in EPA Methd 7471 following each run. The tygon tubing was analyzed as a safety precaution in case there were other forms of Hg, i.e., organic forms such as DMHg, trapped by the tubing. Thus, all forms of Hg being volatilized, including organic forms, would be quantitatively recovered during the anaerobic digestion experiments.

CHAPTER 4
RESULTS

This chapter will be divided into three sections. The first section will present Hg data from Phase I. The second section will present anaerobic reactor data obtained in Phase II and the last section will present Hg data obtained in Phase II.

Hg concentrations in the Alachua County Landfill

In Phase I, the SW composite samples from the Alachua County landfill, which had been split up into fractions as indicated in Chapter 3, were analyzed for total Hg. These fractions had been oven dried and were analyzed for Hg (Hg) using USEPA Method 7471. The Hg data that were obtained for each fraction from the Alachua County landfill are presented in Table 4-1. The fractions are called remaining on 6 mm, remaining on #40, and passing #40. These fractions add up to a total of 100% of dried sample minus what was included in "gross picked" and "fine picked" debris as described in Methods and Materials. Overall, there were 261 fractions for

Table 4-1 Alachua County Landfill Total Mercury Concentrations in three fractions

SAMPLE	DEPTH (m)	Remaining on 8mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g
LFS 1 030	1 52-4.57	66 1	45 1	162
LFS 1 030	4 57 7 62	182	75 8	204
LFS 1 030	7 62-10 67	145	410	368
LFS 1 030	1 52-4 57	407	18 0	76.2
LFS 1 030	4 57-7 62	82.0	78 0	
LFS 1 030	7 62-10 67		131	304
LFS 1 030	10 67-13 72	183	109	447
LFS 1 030	1 52-4 57	88 4	48 1	123
LFS 1 030	4.57-7 62	144	70 6	272
LFS 1 030	7 62-9 14	40 1	68 8	102
LFS 1 030	7 62-10 67			188
LFS 1 040	1 52-4 57	101	48.1	184
LFS 1 040	7 62 10 67	49 8	94 6	315
LFS 1 040	10 67 13 72	118	88 8	208
LFS 1 040	3 05-6.10	96.9		
LFS 1 05			162	
LFS 1 060	0-4 57	491		656
LFS 1 060	4 57 9 14		91 0	236
LFS 1 060	0-4 57	62 8	94.2	129
LFS 1 060	4 57-9 14	293		337
LFS 1 080	0-3 05	267	328	307
LFS 1 080	3 05-6 10	2760	413	2590
LFS 1 080	6 10-9 14	274	267	
LFS 1 090	0-3 05	215	66 7	198
LFS 1 090	3.05-6 10	208	62.4	274
LFS 1 090	6 10-9 14	304	168	228

Table 4-1 (continued)

SAMPLE	DEPTH (m)	Remaining on 6mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g
LFS 1 090	1.52-4.57	263		252
LFS 1 090	4 57 7 62	543		252
LFS 1 090	7 62-10 67	21600		> 70 000
LFS 1 090	10.67 13 72			1650
LFS 1 080	1 52-4.57	245		266
LFS 1 030	4 57 7 62	206		227
LFS 1 080	7 62 10.67	206		678
LFS 1 080	10 67 13 72	313		308
LFS 1 090	1 52-4 57	217		248
LFS 1 080	4 57-7 62	170		358
LFS 1 060	7 62 10.67	268		
LFS 2 030	0-3 05	12 6		
LFS 2 030	3 05-6 10	461	713	2290
LFS 2 030	6 10-9 14	161	418	536
LFS 2 030	0-3 05	12 5	35 1	71 2
LFS 2 030	3 05-6 10	153	30 4	150
LFS 2 030	6 10-9.14	110	97 8	271
LFS 2.060	0-4.57	35 3		
LFS 2 060	4.57 9 14	146	120	185
LFS 2 070	0-3 05		19 1	50 5
LFS 2.070	3.05-6 10		20 3	113
LFS 2.070	6.10-9 14			373
LFS 2.070	7 62-8 38		811	
LFS 2 080	0-3 05	131	66 8	148
LFS 2 080	3 05-6 10	80 2	60.2	139
LFS 2.080	6 10-9 14	566	205	368
LFS 2 080	9 14-12 19	350	209	205
LFS 2 090	12 19-13 72	312	68 7	156
LFS 2 080	0-3 05	67 0	14 8	78 3
LFS 2 080	3 05-6 10	69 3	23 9	103
LFS 2 080	6 10-9 14	461	165	544

Table 4-1 (cont d)

SAMPLE	DEPTH (m)	Remaining on 8mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g
LFS 2 090	0-3.05	17 1		259
LFS 2.090	3 05-6 10	245		561
LFS 2.090	9 10-9 14	483		2110
LFS 2 090	0-3 05	46 4		137
LFS 2 090	3 05-6 10	358		179
LFS 2.090	9 14-12 19	367		860
LFS 2.090	12 19-13 72	85.0		1080
LFS 2,090	9 15-9 14	148		285
LFS 2 090	10 67-13 72	221		190
LFS 3 090	3 05-6 10	1050		426
LFS 3 090	6 10-9 14	623		1110
LFS 3 090	9 14-12.19	39 9		285
LFS 3 090	12 19-13.72	1020		767
LFS 3 090	3.05-6 10	208		558
LFS 3 090	6.10-9 14	1920		606
LFS 3 090	9.14-12 19	1010		3260
LFS 3.090	12 19-13,72	219		1560
LFS 4 090	3 05-6.10	131	46 7	104
LFS 4 090	9 10-9.14	363	60.2	284
LFS 4 090	3 05-6.10	26.5	42.0	46.6
LFS 4 090	6 10-9.14	224	112	214
LFS 5 040	0-3.05	33 9	87 1	144
LFS 5 040	3 05-6 10	119	91 7	124
LFS 5 040	9 10-9 14	761	177	1020
LFS 5 040	9 14-12 19	128	3300	425
LFS 5 040	0-3 05	1270	662	
LFS 5.040	3.05-6 10		122	229
LFS 5 040	9 10-9 14		555	1160
LFS 5 040	9 14-12 19	129	3120	10400
LFS 5.040	0-3-05	33.6	41 4	2 4
LFS 5 040	3 05-6 10	357	329	684
LFS 5 040	9 10-9 14	94 1	162	287
LFS 5 040	9 14-12 19	64 0	102	98 3

Table 4-1 (cont d)

SAMPLE	DEPTH (m)	Remaining on 6mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g
LFS 5 070	1 52-2 29	298	18 3	
LFS 5 070	4 57 5 33			
LFS 5 070	7 62-8 38	247	296	
LFS 5.070	10.67 11 43	176	133	
LFS 5 070	0-3 05		30.5	
LFS 5 070	0-3 05	115	38 5	
LFS 5 070	3 05-6 10	342	211	
LFS 5 070	6 10-9 14	73.5	33 7	
LFS 5.070	9 14 12.19		326	
LFS 5 070	0-3 05		438	925
LFS 5 070	3 05-6.10		269	657
LFS 5 070	6 10-9 14		108	182
LFS 5 070	9 14-12.19		485	480
LFS 5 070	1 52-2.29		65.2	181
LFS 5.070	4.57 5 33		99.8	418
LFS 5 070	7 62-8 38		340	241
LFS 5.070	10 67 11 00		63.8	404
LFS 5 090	1 52-4 57	80 3		188
LFS 5 090	4 57 7 62	195		225
LFS 5 090	7 62-10 67	348		846
LFS 5 090	10 67 13 72	298		677
LFS 5 090	1 52-4 57	97 5		247
LFS 5 090	4 57 7 62	75 2		139
LFS 5 090	7 62-10 67	674		2500
LFS 5 090	10 67 13 72	251		318

All Raw Data are presented in Appendix A.
Spike recoveries were within limits (80 - 120%)
R^2 values for standard curves were > 0 990

which total Hg was determined. The total Hg that was computed in the various fractions ranged from 12.5 to greater than 70,000 ng/g Hg. Raw data along with AQ/QC for Phase 1 are shown in Appendix A.

It should be noted that in the digestion step, an excess of potassium permanganate ($KMnO_4$), one of the oxidizers, had to be added so that the purple color would persist in the sample solution. The $KMnO_4$ that was required was 30 45 mL versus the required 15 mL in the USEPA Method.

After total Hg was determined in each of the three fractions, total Hg in the composite samples was calculated by multiplying the percent of the composite weight that each fraction represented by the total Hg concentration in that fraction and adding the three numbers together. This gives the Hg (ng/g) in the composite sample, i.e.

$$\text{Hg in Composite} = \sum F_i C_i / 100$$

where, F_i is the weight percent of one of the three fractions, C_i is the Hg concentration corresponding to the weight fraction i, and i goes from 1 to 3. Table 4-2 shows the percentages that each fraction comprised of the total composite sample weight, F_i. Values for C_i were taken from Table 4-1. Table 4-3 shows the Hg concentrations in the

Table 4 2 % Dry weight of Total Composite Sample

SAMPLE	Remaining on 5 mm	Passing #40	Remaining on #40
LFS 1.0301	46.0	36.5	17 1
LFS 1 0301	40 1	48 1	11 8
LFS 1 0301	38 5	44.3	17 2
LFS 1 0302	41 8	51 3	6.5
LFS 1 0302	41 8	46 3	11 5
LFS 1 0302	34 9	49 4	15 7
LFS 1 0302	25 3	58.9	15 8
LFS 1 0303	52 2	33.4	14 4
LFS 1 0303	38 8	47 7	13.5
LFS 1 0303	31 6	48 3	20 1
LFS 1 0303			
LFS 1 0401	25.9	53.9	20 2
LFS 1 0401	17 0	66 3	16 7
LFS 1 0401	25 7	63 3	11 0
LFS 1 0402			
LFS 1 06	18 7	65.5	15 8
LFS 1 0602	31 0	46 6	22 4
LFS 1 0602	21 1	53 6	25 3
LFS 1 0603	39 9	22.2	37 9
LFS 1 0603	14 3	22 0	63 7
LFS 1 0601	35 1	54 4	10 4
LFS 1 0801	35 1	52.2	12.6
LFS 1 0801	34 7	52 1	13 3
LFS 1 0802	64 0	22 7	13 3
LFS 1 0802	29 7	60 2	10.2
LFS 1 0802	39 1	44.3	16 6

Table 4.2 (Cont'd)

SAMPLE	Remaining on 6 mm	Passing #40	Remaining on #40
LFS 1.0901	35.1	54.4	10.4
LFS 1.0901	35.1	52.2	12.6
LFS 1.0901	34.7	52.1	13.3
LFS 1.0901	64.0	22.7	13.3
LFS 1.0902	28.7	60.2	11.2
LFS 1.0902	39.1	44.3	16.6
LFS 1.0902	43.8	44.6	11.6
LFS 1.0902	43.5	49.0	7.6
LFS 1.0903	35.3	52.5	12.3
LFS 1.0903	39.1	44.4	16.5
LFS 1.0903	44.7	39.0	16.3
LFS 2.0301	100.0		
LFS 2.0301	38.1	51.1	10.8
LFS 2.0301	22.6	59.6	17.7
LFS 2.0302	29.7	57.9	12.4
LFS 2.0302	31.0	61.0	8.0
LFS 2.0302	42.6	36.6	20.8
LFS 2.0603	43.3	29.5	27.2
LFS 2.0603	21.1	60.4	18.5
LFS 2.0701	33.5	52.1	14.4
LFS 2.0701	25.8	62.8	11.4
LFS 2.0701	33.9	49.1	17.0
LFS 2.0702			
LFS 2.0801	43.6	44.6	11.8
LFS 2.0801	43.5	49.0	7.6
LFS 2.0801	35.3	52.5	12.3
LFS 2.0801	39.1	44.4	16.5
LFS 2.0801	44.7	39.0	16.3
LFS 2.0802	49.8	37.7	12.5
LFS 2.0802	47.0	42.2	10.8
LFS 2.0802	25.6	64.7	9.8

Table 4-2 (Cont'd)

SAMPLE	Remaining on 6 mm	Passing #40	Remaining on #40
LFS 2 0901	47.0	42.2	10.8
LFS 2 0901	25.6	64.7	9.8
LFS 2 0901	16.8	74.6	8.6
LFS 2 0902	26.0	62.3	11.7
LFS 2 0902	18.4	74.6	6.9
LFS 2 0902	54.6	26.7	18.8
LFS 2 0902	52.6	16.3	17.9
LFS 2 0903	46.1	37.4	16.5
LFS 2 0903	26.5	58.3	16.2
LFS 3.0901			
LFS 3.0901			
LFS 3 0901			
LFS 3.0901			
LFS 3 0902			
LFS 3 0902			
LFS 3 0902			
LFS 3 0902			
LFS 4.0801	16.8	74.6	8.6
LFS 4 0801	26.0	62.3	11.7
LFS 4.0802	16.4	8.6	74.6
LFS 4 0802	30.6	14.6	54.8
LFS 5 0401	42.1	40.2	17.7
LFS 5 0401	56.6	26.0	17.6
LFS 5 0401	56.3	30.0	13.7
LFS 5 0401	16.6	62.5	18.7
LFS 5.0402	66.4	20.1	13.5
LFS 6 0402	18.5	65.9	16.5
LFS 6 0402	41.6	36.0	18.5
LFS 3 0402	32.7	53.2	14.1
LFS 6 0403	29.6	58.7	11.7
LFS 6 0403	63.0	22.3	14.7
LFS 5 0403	63.4	22.3	14.3
LFS 5 0403	33.1	45.7	21.2

Table 4-2 (Cont'd)

SAMPLE	Remaining on 5 mm	Passing #40	Remaining on #40
LFS 5.0701	24.1	70.4	5.5
LFS 5.0701			
LFS 5.0701	71.3	12.2	16.5
LFS 5.0701	79.0	11.1	9.9
LFS 5.0702			
LFS 5.0703	33.4	55.5	11.1
LFS 5.0703			
LFS 5.0703	53.8	29.0	17.2
LFS 5.0703	41.9	32.7	25.4
LFS 5.0704	48.6	37.5	13.9
LFS 5.0704			
LFS 5.0704	58.7	29.5	11.8
LFS 5.0704	56.5	18.5	25.1
LFS 5.0705	25.4	67.5	7.1
LFS 5.0705	22.8	61.5	15.7
LFS 5.0705	71.8	17.1	11.1
LFS 5.0705	16.1	73.7	10.3
LFS 5.0801			
LFS 5.0801			
LFS 5.0801			
LFS 5.0801			
LFS 5.0802			
LFS 5.0802			
LFS 5.0802			
LFS 5.0802			

Table 4-3. Alachua County Landfill Mercury Concentrations[21] in the composite (weighted sum of the 3 fractions)

SAMPLE	DEPTH (m)	Remaining on 6 mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g	Composite [Hg] ng/g
LFS 1 0301	1 52-4 57	31.8	18 8	27 7	76 2
LFS 1 0301	4 57 7 62	65 0	36 3	24 1	125
LFS 1 0301	7 62 10.67	56 9	182	63 3	301
LFS 1 0302	1.52-4 57	171	6 2	8 2	156
LFS 1 0302	4 57 7 62	34 3	36 6	8 0	70 8
LFS 1 0302	7 62 10 67		64 8	47 8	112
LFS 1 0302	10 67-13.72	46.3	64.2	71 0	182
LFS 1 0303	1 52-4 57	35 7	15 1	17.8	68 8
LFS 1 0303	4 57 7 62	55 8	33.7	36 8	128
LFS 1 0303	7 62-9 14	12 7	32,3	20 4	65.4
LFS 1 0303	7 62-10 67				
LFS 1 0401	1 52-4 57	26 3	28 8	39 2	91.4
LFS 1 0401	7 62-10 67	8 8	55 1	52.8	117
LFS 1 0401	10 67-13 72	29 8	81 8	22.8	114
LFS 1 0402	3 05-6 10				
LFS 1 05			107		107
LFS 1 0602	0-4 57	152		181	343
LFS 1 0602	4.57 9 14		46.8	60,5	106
LFS 1 0603	0-4 57	25 1	14.2	48 0	68 3
LFS 1 0603	4 57-9.14	41 8		218	257
LFS 1 0801	0-3 05	93 8	179	32 0	305
LFS 1 0801	3 05-6 10	367	218	330	1510
LFS 1 0801	6.10-9 14	95 0	139		234
LFS 1 0802	0-3 05	138	18 7	26 3	184
LFS 1 0802	3 05-6 10	81 8	37.5	27 8	127
LFS 1 0802	6 10-9 14	118	74.6	36 4	236

Table 4-3 (cont'd)

SAMPLE	DEPTH (m)	Remaining on 6 mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g	Composite [Hg] ng/g
LFS 1 0901	1.52-4.57	92.8		28.3	116
LFS 1 0901	4.57-7.62	190		32.1	222
LFS 1 0901	7.62-10.67	7470		9280	16800
LFS 1 0901	10.67-13.72			218	218
LFS 1 0902	1.52-4.57	72.6		30.4	103
LFS 1 0902	4.57-7.62	80.7		37.6	118
LFS 1 0902	7.62-10.67	91.2		68.6	159
LFS 1 0902	10.67-13.72	136		23.1	159
LFS 1 0903	1.52-4.57	76.4		30.6	107
LFS 1 0903	4.57-7.62	66.5		58.2	126
LFS 1 0903	7.62-10.67	120			120
LFS 2 0301	0-3.05	12.6			12.5
LFS 2.0301	3.05-6.10	176	366	247	789
LFS 2.0301	6.10-9.14	40.6	248	96.5	385
LFS 2 0302	0-3.05	3.7	20.3	8.8	32.8
LFS 2 0302	3.05-6.10	47.4	18.6	12.0	77.6
LFS 2.0302	6.10-9.14	46.7	36.0	55.6	138
LFS 2 0603	0-4.57	15.3			15.3
LFS 2 0603	4.57-9.14	31.2	72.7	34.2	138
LFS 2.0701	0-3.05		6.8	7.3	14.1
LFS 2.0701	3.05-6.10		12.8	12.9	25.7
LFS 2.0701	6.10-9.14		0.0	63.4	63.4
LFS 2 0702	7.62-9.38				
LFS 2 0801	0-3.05	57.2	30.0	17.2	104
LFS 2 0801	3.05-6.10	34.9	29.5	10.4	74.8
LFS 2 0801	6.10-9.14	207	108	45.2	360
LFS 2 0801	9.14-12.19	137	80.6	33.6	252
LFS 2 0801	12.19-13.72	136	26.6	26.0	162
LFS 2.0802	0-3.05	33.4	5.6	8.8	46.7
LFS 2.0802	3.05-6.10	42.0	10.1	11.1	63.2
LFS 2 0802	6.10-9.14	123	127	53.1	303

Table 4-3 (cont'd)

SAMPLE	DEPTH (m)	9 mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g	Composite [Hg] ng/g
LFS 2.0901	0-3.05	8.1		27.9	36.0
LFS 2.0901	3.05-6.10	62.9		54.7	119
LFS 2.0901	6.10-9.14	80.0		182	262
LFS 2.0902	0-3.05	12.1		16.1	28.2
LFS 2.0902	3.05-6.10	58.9		15.7	74.2
LFS 2.0902	9.14-12.19	200		145	345
LFS 2.0902	12.19-13.72	82.2		193	255
LFS 2.0903	6.10-9.14	66.7		48.7	115
LFS 2.0903	10.67-13.72	85.2		28.9	54.2
LFS 3.0901	3.05-6.10				
LFS 3.0901	6.10-9.14				
LFS 3.0901	9.14-12.19				
LFS 3.0901	12.19-13.72				
LFS 3.0902	3.05-6.10				
LFS 3.0902	6.10-9.14				
LFS 3.0902	9.14-12.19				
LFS 3.0902	12.19-13.72				
LFS 4.0901	3.05-6.10	21.7	34.8	9.9	65.8
LFS 4.0901	6.10-9.14	94.3	37.5	33.2	168
LFS 4.0902	3.05-6.10	4.7	3.7	36.8	44.9
LFS 4.0902	6.10-9.14	89.7	19.2	118	203
LFS 5.0401	0-3.05	14.3	39.0	25.4	78.7
LFS 5.0401	3.05-6.10	64.7	19.3	21.9	102
LFS 5.0401	6.10-9.14	440	53.1	139	632
LFS 5.0401	9.14-12.19	24.3	2060	79.4	2160
LFS 5.0402	0-3.05	945	103		1098
LFS 5.0402	3.05-6.10		80.9	35.5	116
LFS 5.0402	6.10-9.14		217	228	443
LFS 5.0402	9.14-12.19	42.3	1660	1860	3160
LFS 5.0403	0-3.05	8.6	24.7	9.3	34.8
LFS 5.0403	3.05-6.10	225	73.2	130	428
LFS 5.0403	6.10-9.14	56.7	36.1	41.1	137
LFS 5.0403	9.14-12.19	21.2	46.5	20.9	89.5

Table 4-3 (cont'd)

SAMPLE	DEPTH (m)	6 mm [Hg] ng/g	Passing #40 [Hg] ng/g	Remaining on #40 [Hg] ng/g	Composite [Hg] ng/g
LFS 5.0701	1.52-2.29	71.6	11.5		83.1
LFS 5.0701	4.57-5.33				
LFS 5.0701	7.62-8.38	178	39.6		212
LFS 5.0701	10.67-11.43	139	14.8		154
LFS 5.0702	0-3.05				
LFS 5.0703	0-3.05	38.3	20.3		58.6
LFS 5.0703	3.05-6.10				
LFS 5.0703	6.10-9.14	39.6	9.8		49.3
LFS 5.0703	9.14-12.19		107		107
LFS 5.0704	0-3.05		164	129	293
LFS 5.0704	3.05-6.10		256	667	616
LFS 5.0704	6.10-9.14		106	182	270
LFS 5.0704	9.14-12.19		485	460	925
LFS 5.0705	1.52-2.29		44.0	12.6	56.6
LFS 5.0705	4.57-5.33		61.3	65.6	127
LFS 5.0705	7.62-8.38		58.1	28.7	84.8
LFS 5.0705	10.67-11.00		47.0	41.4	88.4
LFS 5.0901	1.52-4.57				
LFS 5.0901	4.57-7.62				
LFS 5.0901	7.62-10.67				
LFS 5.0901	10.67-13.72				
LFS 5.0902	1.52-4.57				
LFS 5.0902	4.57-7.62				
LFS 5.0902	7.62-10.67				
LFS 5.0902	10.67-13.72				

Composite = $\Sigma F_i C_i$

where: F_i = weight % of one of the 3 size fractions

C_i = Hg concentration corresponding to F_i

composite samples. The total Hg in the composite samples ranged from 32.8 to 16800 (ng/g Hg).

Total Hg determinations were performed to quantify the amount of Hg in solid waste samples from the Alachua County Landfill which had received MSW from 1973 to the present. Mercury was detected in every sample analyzed. "Hot spots" exist in the Alachua County Landfill which probably result from the decomposition of discrete Hg-containing devices such as batteries and light bulbs that are still finding their way into the landfill today in spite of more stringent recycling efforts.

Mercury concentrations in Palm Beach County compost samples are shown in Table 4-4. The values ranged from 368 ng/g Hg to 5320 ng/g Hg with a mean of 1100 ng/g Hg. The mean Hg concentration for the compost samples was higher than that for the MSW samples from the Alachua County landfill and the distribution of concentrations exhibited a much narrower range.

Phase II-Anaerobic Reactor Data

Results from Phase I established that Hg did in fact exist in the Alachua County landfill. The next step was to evaluate what happens to Hg during anaerobic digestion like

Table 4-4 Palm Beach County Mercury Data (1:1 Biosolids to Yard Waste)

Sample	Hg (ng/g)
1	733
2	514
3	930
4	635
5	405
6	420
7	550
8	854
9	531
10	372
11	355
12	5320
13	1260
14	1680
15	604
16	368
17	450
18	591
19	1580
20	2770
21	1700
22	1610
23	433
24	416
average	1100

that occurring in the Alachua County landfill Data for the simulated landfill Run 1 are presented in Figures 4-1 to 4-12 Run 2 data are presented in Figures 4-13 to 4-28 These figures show that the pH's, volatile fatty acids, methane, and methane yield acted as expected under anaerobic conditions The acid-producing microbes were active around day 5 in both runs for all of the reactors as indicated in the pH graphs The pH was lowered to less than 7 at day 5, but increased to alkaline levels after day 5 The methane content initially started around zero in all cases because at the start of the runs mostly nitrogen and oxygen were present until anaerobic conditions were established The methane level steadily increased from day 1 until it leveled off at around 55% The volatile fatty acids (VFAs) were indicative of the action of the microbes in the system and at day 5 it was evident that there was an increase in the VFA concentrations which then decreased after that time

The methane yield figures are the most important determinants in establishing that the reactors were operating under anaerobic conditions A document by Chynoweth et al. (1991) provided data on ultimate methane yield values for newspaper and office paper. The value given for newspaper was

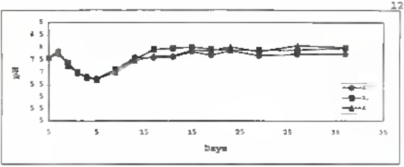

Figure 4-1 Run 1 (Control) pH vs Days

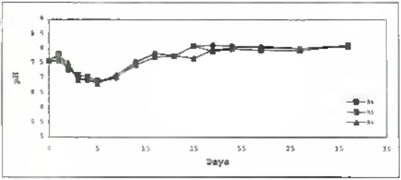

Figure 4-2 Run 1 (100ng Hg) pH vs Days

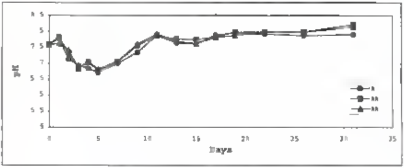

Figure 4 3 Run 1 (2000ng Hg) pH vs Days

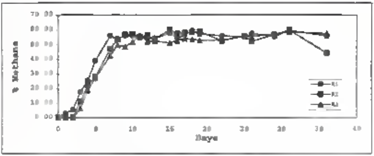

Figure 4.4 Run 1 (Control) %Methane vs. Days

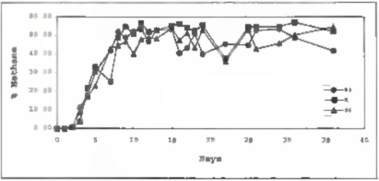

Figure 4.5 Run 1 (100mg Mg) %Methane vs. Days

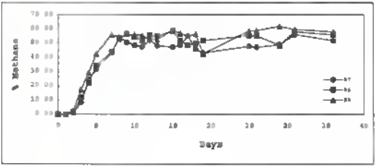

Figure 4.6 Run 1 (2000mg Mg) %Methane vs. Days

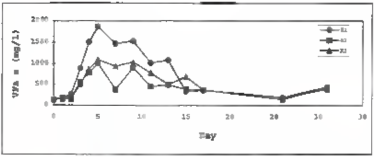

Figure 4-7 Run 1 (Control) Volatile Fatty Acids vs Days

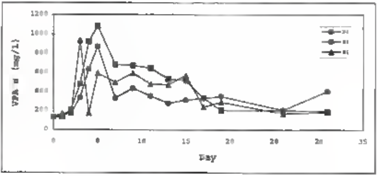

Figure 4-8 Run 1 (100mg Hg) Volatile Fatty Acids vs Day

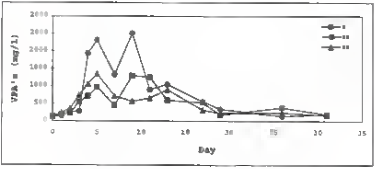

Figure 4-9 Run 1(2000mg Hg) Volatile Fatty Acids vs Day

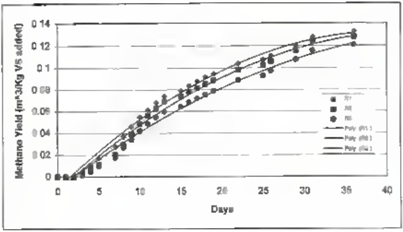

Figure 4-10 Run 1 (Control) Methane Yield vs Days

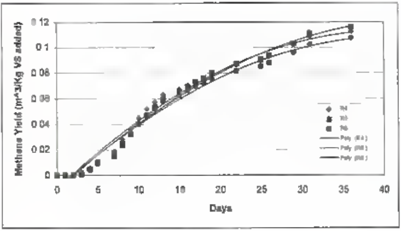

Figure 4-11 Run 1 (100 ng Mg) Methane Yield vs Days

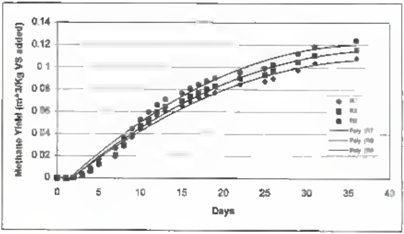

Figure 4-12 Run 1 (2000 ng Hg) Methane Yield vs Days

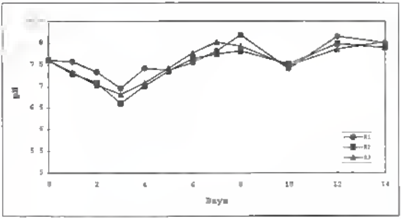

Figure 4 13 Run 2 (Control) pH vs Days

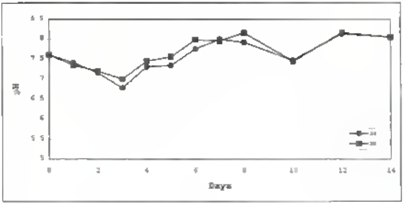

Figure 4-14 Run 2 (100ng Hg) pH vs Days

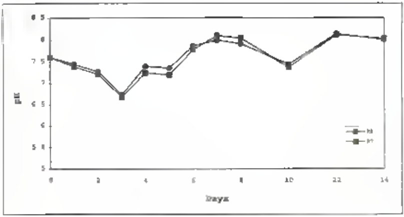

Figure 4-15 Run 2 (1000mg Hg) pH vs. Days

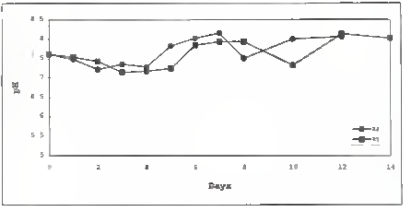

Figure 4-16 Run 2 (2000mg Hg) pH vs. Days

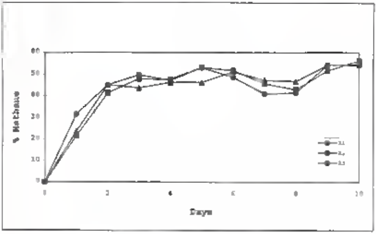

Figure 4.17 Run 2 (Control) % Methane vs Days

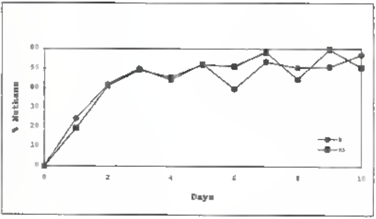

Figure 4.18 Run 2 (100ng Hg) % Methane vs Days

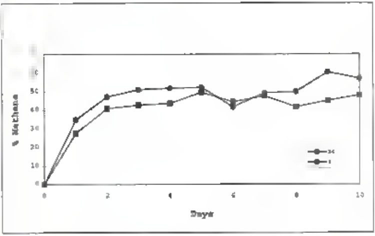

Figure 4-19 Run 2 (1000ng Hg) % Methane vs Days

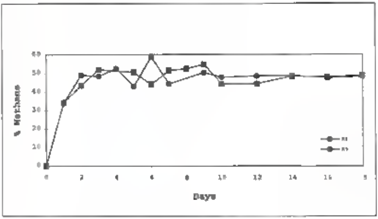

Figure 4-20 Run 2 (2000ng Hg) % Methane vs Days

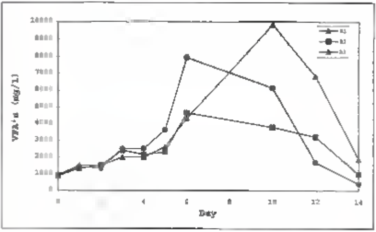

Figure 4-21 Run 2 (Control) Volatile Fatty Acids vs Days

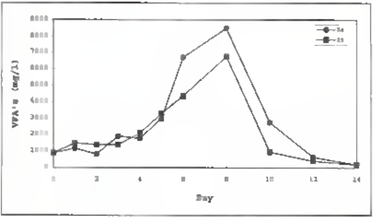

Figure 4-22 Run 2 (100mg Mg) Volatile Fatty Acids vs Days

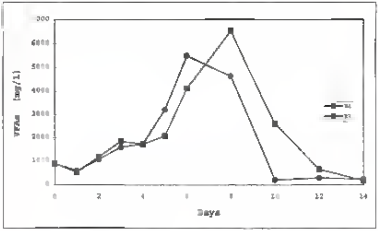

Figure 4-23 Run 2 (1000mg Mg) Volatile Fatty Acids vs Days

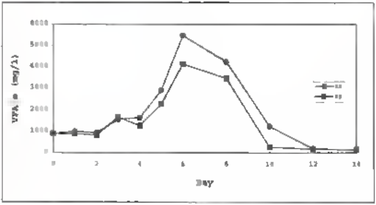

Figure 4-24 Run 2 (2000mg Mg) Volatile Fatty Acids vs Days

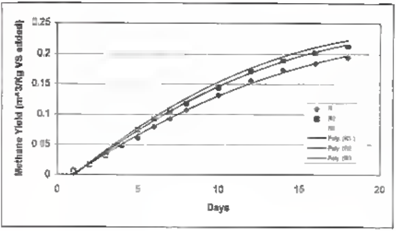

Figure 4-25 Run 2 (Controls) Methane Yield vs Days

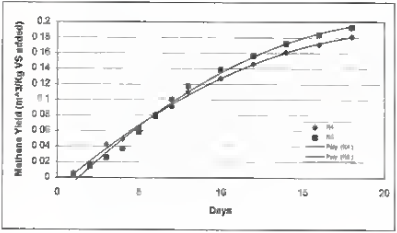

Figure 4-26 Run 2(100 ng Hg) Methane Yield vs Days
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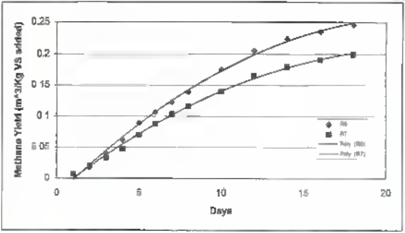

Figure 4-27 Run 2(1000 ng Hg) Methane Yield vs Days

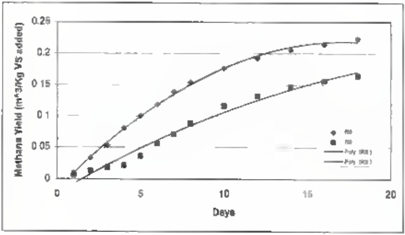

Figure 4-28 Run 2(2000 ng Hg) Methane Yield vs Days

0.100 m^3/Kg "VS added" and 0.369 m^3/Kg "VS added" for office paper. Since 90% of the SW in the reactors consisted of newspaper and office paper, these are the values that were used for the determination of the expected yields of the reactors. The methane yield for all of the reactors fell within the range 0.100 - 0.369 m^3/Kg "VS added". The methane yield results for Run 2 were higher than that for Run 1 and this is attributed to silicone being used as a sealant along with the teflon tape. In Run 1 only teflon tape was used. This section includes all of the reactor data excluding the Hg. The raw data are provided in Appendix B.

Phase II-Mercury Data

The raw data from the total Hg analyses performed on the solid, liquid, and gas phases from the reactors are presented in Appendix C. The solid phase was the SW, the liquid phase was the leachate, and the gas phase included the contributions from the activated carbon traps and the tygon tubing leading from the reactors to the carbon traps. Table 4-5 shows a compilation of the Hg mass balances. The data from the individual runs are presented in Figures 4-29 to 4-35.

In doing the Hg mass balances, it must be noted that most of the levels recovered did not add up to 100% of the Hg that

Table 4.5 Mercury Recovered

Run	Reactor	Total Hg Added (ng)	Solids* (ng)	Leachate (ng)	Gms Act C (ng)	Gms Tygon (ng)	Percent Recovery
1	1	0	8**	0	0	0	
	2	0	8	0	0	0	
	3	0	4	0	0	0	
	4	100	55	12	18	12	97.0
	5	100	53	0	8	28	81.0
	6	100	67	0	18	16	101.0
	7	2000	1760	0	24	26	90.5
	8	2000	1480	0	30	18	76.4
	9	2000	1910	0	34	8	97.2
2	1	0	8	0	0	0	
	2	0	9	0	0	0	
	3	0	10	0	0	0	
	4	100	65	0	25	18	108.0
	5	100	47	0	32	16	95.0
	6	1000	836	28	90	28	98.2
	7	1000	810	0	75	8	88.5
	8	2000	1890	0	30	18	96.9
	9	2000	1810	0	132	4	97.1

* Does not include 6 - 15 grams of dry weight that was removed

** Method Detection Limit for all analyses was 8 ng Hg

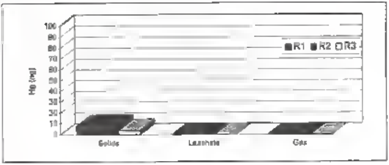

Figure 4-29 Run 1 Hg content in 3 phases (control)

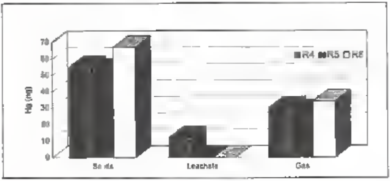

Figure 4-30 Run 1 Hg content in 3 phases (100 ng Hg)

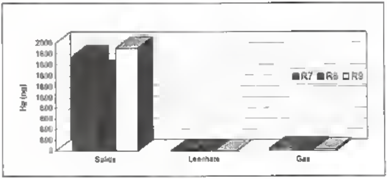

Figure 4-31 Run 1 Hg content in 3 phases (2000 ng Hg)

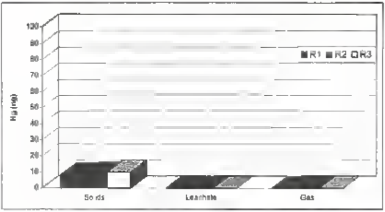

Figure 4-32 Run 2 Hg content in 3 phases (control)

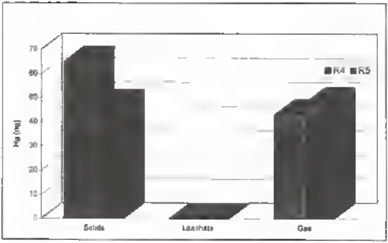

Figure 4-33 Run 2 Hg content in 3 phases (100 ng Hg)

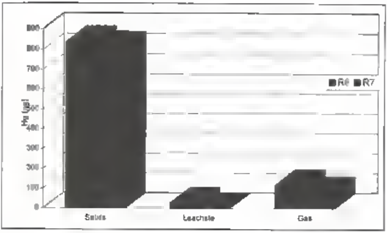

Figure 4-34 Run 2 Hg content in 3 phases (1000 ng Hg)

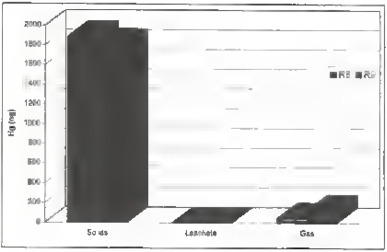

Figure 4-35 Run 2 Hg content in 3 phases (2000 ng Hg)

was originally spiked into the reactors. Part of this can be attributed to experimental error. Another part can be attributed to the SW that was removed from the reactors after the completion of the run for the purpose of volatile solids analysis.

Run 1. In Run 1, reactors 1, 2, and 3 served as the control reactors and did not receive any Hg. No Hg was detected in either the leachate or gas phase of the control reactors. However, low levels of Hg (8, 3, and 4 ng) were observed in the SW itself, but none of these values exceeded the MDL of 8 ng.

Reactors 4, 5, and 6 were spiked with 100 ng of Hg and 30, 36, and 34 ng Hg, respectively, were found in the carbon traps and tygon tubing leading to the traps. This accounted for 30%, 36%, and 34% of the Hg added being volatilized. Twelve ng Hg were detected in the leachate in reactor 4, but none was found in reactors 5 and 6. The rest of the Hg was detected in the solid phase, i.e., 55%, 53%, and 67%, respectively. The recovery of Hg was 97, 89, and 101% for reactors 4, 5, and 6, respectively, not counting that in the 6 g of SW that had been removed for volatile solids analysis.

Reactors 7, 8, and 9 were spiked with 2000 ng of Hg and 50, 48, and 42 ng Hg, respectively, were found in the carbon traps and tygon tubing. This accounted for 2.5%, 2.4%, and 2.1% of the added Hg being volatilized. There was no Hg detected in the leachate in any of these reactors. The Hg detected in the SW was 1760, 1480, and 1910 ng Hg for reactors 7, 8, and 9, respectively. This accounted for 88%, 74% and 95% of the total Hg added, again not including the 6 g of SW that were removed for volatile solids analysis.

Run 2. Reactors 1, 2, and 3 again served as controls and received no Hg. In these reactors, there was no Hg detected in the gas or leachate phases, although low levels were again detected in the SW, but the values were at or below the MDL being 8, 9, and 10 ng for reactors 1,2, and 3, respectively.

Reactors 4 and 5 were spiked with 100 ng Hg and 37 and 48 ng Hg, respectively, were found in the carbon traps and tygon tubing. This accounted for 37% and 48% of the Hg added being volatilized. No Hg was detected in either of the leachates. The rest of the Hg was detected in the solid phase, i.e., 61% and 47% respectively. The recovery of Hg was 108 and 95% for reactors 4 and 5, respectively, not including 10 g of SW that was removed for volatile solids analysis. The distribution of

Hg between solid and gaseous phases was similar to that observed in Run 1

Reactors 6 and 7 each received 1000 ng of Hg and 128 and 83 ng Hg respectively were found in the carbon traps and tygon tubing This accounted for 12 8% and 8 3% of the added Hg being volatilized Reactor 6 contained 28 ng Hg in the leachate but reactor 7 did not show any Hg in the leachate The Hg detected in the SW was 836 and 810 ng Hg for reactors 6 and 7 respectively This accounted for 84% and 81% of the total Hg added The recovery of Hg was 98 and 89% again not including the 10 g of SW removed for volatile solids analysis

Reactors 8 and 9 each received 2000 ng of Hg and 48 and 138 ng Hg respectively were found in the carbon traps and tygon tubing These levels represented 2 4% and 6 6% of the total Hg being volatilized There was no Hg detected in the leachate of either reactor Reactor 8 contained 1890 ng Hg in the SW which was 94% of the total Hg added and reactor 9 had 1810 ng Hg which was 90% of the total The mass balance was 97% for both reactors, not including that in the 10 g of SW that was taken for volatile solids analysis

CHAPTER 5
DISCUSSION

Phase I - Hg in the Alachua County Landfill

In the Phase I experiments, the data showed that there was Hg in the Alachua County Landfill. The average concentration for composite samples was 176 ng/g with over half of the values being below 150 ng/g. Figure 5.1 shows the frequency distribution of Hg obtained in composite samples from the Alachua County Landfill. From the figure, it is evident that most of the samples lie within 100-150 ng/g with fewer samples appearing at the other concentrations. However, five values exceeded 1000 ng/g with one exceeding 16,000 ng/g. These values are important in that they are considerably higher than background Hg concentrations in Florida soils which range from 0.5 ng/g to 43 ng/g (Ma et al. 1997). However, they are considerably lower than current target clean-up levels set by Florida DEP for soil. The target value for Hg in residential soil is 23,000 ng/g and in industrial

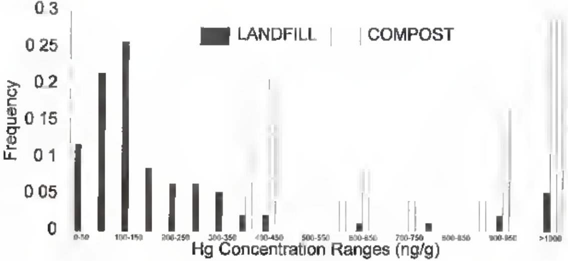

Figure 5.1 Hg conc ranges(ng/g) in Alachua County Landfill & Palm Beach County compost

soil is 480,000 ng/g (Florida Department of Environmental Protection, 1995). The maximum concentration for Hg in sewage sludge as set forth in CFR 40 Part 503 is 57,000 ng/g (Code of Federal Regulations, 1997). Thus, if the Alachua County Landfill were to be reclaimed and the SW residue taken out and land applied, it would meet requirements for Hg that are much more stringent than is currently required for sewage sludge. Palm Beach County compost data is also included in Figure 5-1. This compost consisted of a 50:50 mixture of biosolids to yard waste. These are consistently higher than the MSW Hg data, but are still well below the regulatory limits.

LFS 1, LFS 2 and LFS 5 are sites on a fairly new portion of the landfill and therefore might be expected to contain lower levels of Hg due to modern recycling efforts. A comparison of Hg concentrations found in LFS 2 and LFS 4, which differed only in age and the presence of a liner, showed that Hg concentrations were actually lower at LFS 4 than at LFS 2 but the differences were not significant at the 5% probability level. LFS 4 received MSW beginning in 1973 while LFS 2 began receiving MSW in 1989, the year that disposal of batteries in MSW landfills was prohibited. Although there have been recycling efforts for household batteries that contain Hg since 1988. Hg is still making its way into landfills via other sources such as Hg light switches, electric lighting, paint residues, fever thermostats, pigments, dental uses and special paper coating (Bureau of Solid and Hazardous Waste 1995). Since Hg is present in MSW from these sources there is a need to better understand the fate of this Hg.

The Hg levels determined in the SW fractions in Table 4-1 ranged from 12.5 ng/g to greater than 70,000 ng/g Hg. The Hg levels in the composite samples ranged from 32.8 ng/g to 16,600 ng/g Hg. Table 4-2 is missing Hg concentration data for some composite samples. These missing data resulted from the fact that dry weight data for some size fractions were not available. In addition, for some core samples, one or more fractions were missing and could not be analyzed for Hg. However, in spite of the missing fractions and weight data, results for composite samples were calculated for 53 core samples. This number should be sufficient to say something about Hg concentrations in the Alachua County Landfill. It is assumed that these concentrations would also be representative of the Hg concentrations that exist in landfills for other municipalities around the country. Unfortunately, a search of

The literature did not yield Hg data from other landfills to allow validation of this assumption

Samples were taken from all five areas of the landfill i.e. LFS 1 to LFS 5. LFS 1 and LFS 2 were among the more recent additions to the landfill, with LFS 1 having leachate recycling and LFS 2 acting as a control without leachate recycling Thus a comparison of Hg concentrations in LFS 1 and LFS 2 would show the effect of leachate recycling As a result of leachate recycling, LFS 1 would have enhanced microbial activity due to the higher moisture content than would LFS 2 The enhanced microbial activity would accelerate the degradation rate of the SW. Differences between LFS 1 and LFS 2 were evaluated at each depth and the results are shown in Table 5-1 It must be noted that the 0 - 3 05 m (0 - 10 ft) depth in LFS 2 was equivalent to the 1.52 - 4 57 m (5 15 ft) depth in LFS 1 with the other depths corresponding in like fashion The landfill workers had added fresh material to LFS 1 after the sampling had started and markers were placed so the first 1 52 m were discarded in order to be consistent with the depths in LFS 2.

The overall means for LFS 1 (155 ng/g) and LFS 2 (153 ng/g) were almost identical and not significantly different

Table 5-1 Mean Hg concentrations in composite samples for LFS 1 and LFS 2 Alachua County Landfill

Depth (m)	LFS 1 (ng/g Hg)	LFS 2 (ng/g Hg)
0 - 3 05	152 (11)[1]	36 (8)*
3 05 - 6 10	142 (9)	174 (7)
6 10 - 9 14	167 (8)	221 (8)
9 14 - 12 19	168 (4)	234 (3)
Overall(w/in a row)	155 (32)	153 (26)

* Means within a row are significantly different at the 1% probability level based on student's two tailed t test

[1] Number in parentheses are the number of observations for that treatment

Thus leachate recycling did not effect the total amount of Hg remaining in the solid residues. Since LFS 1 and LFS2 were both from lined areas, the only way that they could differ in Hg concentration is if Hg volatilization was different. Thus, leachate recycling did not appear to affect Hg volatilization. However, leachate recycling did affect the distribution of Hg with depth. At LFS 1, the Hg concentrations were fairly consistent with depth, ranging from 152 ng/g in the top layer to 168 ng/g Hg in the bottom layer. These differences were not significant (Fig. 5-2). Thus, leachate recycling resulted in a more even distribution of Hg from the top layer (1.52 - 4.57 m) to the bottom layer (10.67 - 13.72 m).

Confidence intervals (95%) were calculated using the formula

$$\text{C.I. } (95\%) = \pm\, t_{0.05}\, \text{s.e.}$$

where $t_{0.05}$ represents the two-tailed test at a probability value of 0.05 for a given value of degrees of freedom. The $t_{0.05}$ values were obtained from Snedecor and Cochran (1968). The s.e. represents the standard error and this is determined using the standard deviation divided by the degrees of freedom. The error bar on the 9.14 - 12.19 m depth for LFS 2 (Fig. 5-1) is very large for two reasons. First, the mean

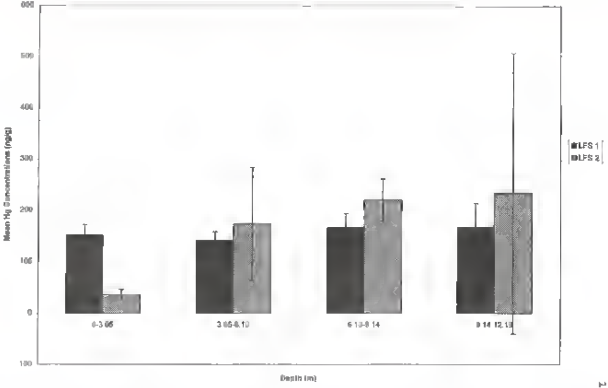

Figure 5.2 Mean Hg concentrations with depth for LFS1 and LFS2

consisted of only three observations, yielding a small number of degrees of freedom and a relatively high $t_{0.05}$ value. Second, the three observations ranged from 94 to over 300 ng/g, resulting in a large standard deviation.

At LFS 2, there was a trend of increasing Hg concentration with increasing depth. The averages that were computed for the depths also exhibited a considerable range, i.e. 36.5 ng/g at the top to 234 ng/g Hg at the bottom. Confidence intervals (95%) for each depth (Figure 5-2) showed that the top layer (0 – 3.05 m) at LFS 2 was significantly lower in Hg than the lower three depths. The lower three depths did not differ significantly in their Hg concentrations. The trend of increasing Hg concentrations with depth was also evident at LFS 3 and LFS 4 where Hg concentrations two to three times higher than that in the top layer could be found in lower layers (Table 4-2). This suggests that, in the absence of leachate recycling, Hg tends to leach and settle in the deepest layers. Leachate recycling may be beneficial in evenly distributing Hg, and maybe other heavy metals within the residue, so that the highest concentrations are not at the lowest layers. These data also suggest that Hg does tend to leach downward through a landfill and that soil and groundwater contamination with Hg could pose a threat in the case of unlined landfills.

Statistical comparisons were also performed for the 3.05-9.14 m depth at LFS 2 and LFS 4 (Table 5-2). This was the only depth for which Hg data were available at LFS 4. These two sites (LFS 2 and LFS 4) differed with respect to age and neither was involved with leachate recycling. LFS 4 is the oldest site at the landfill and had been in operation since 1973. It was unlined, whereas LFS 2, a site that had been receiving MSW since 1988, was lined. Thus, in addition to volatile losses that could occur at both sites, LFS 4 could also have lost Hg by leaching over the years it had been in use. LFS 4 was capped in 1985 to prevent leaching losses, but was open from its initiation in 1973 until then. A comparison of LFS 2 and LFS 4 would provide some indication of the effect of the age of the landfill with the Hg concentrations.

The Hg concentrations in the composite samples at LFS 4 were lower than those at LFS 2. However, a t-test showed that the difference between the two means in this table was not significant. In a recent review of leachate attenuation at landfills, Christensen et al. (1994), concluded that heavy metals, including Hg, do not constitute a significant

Table 5-2 Mean Hg concentrations in composite samples for LFS 2 and LFS 4 Alachua County Landfill

Depth (m)	LFS 2 (ng/g Hg)	LFS 4 (ng/g Hg)
3 05 - 9 14	200 (4)¹	120 (4)*

* Means were not significantly different at the 5% probability level based on student's two-tailed t-test

¹ Number in parentheses are the number of observations for that treatment

pollution problem at landfills due to low concentrations in the leachate and strong attenuation by sorption and precipitation. If this is true, then leaching would not be a factor in this comparison. Thus, any differences in Hg concentration in the solid residue could be attributed to differences in Hg volatilization. Assuming similar volatilization losses between LFS 2 and 4, the data suggest that the implementation of recycling laws has had little impact on Hg concentrations in the Alachua County Landfill. This may result from the fact that many Hg-containing devices are still making their way into landfills.

Hg concentrations were computed for the fractions remaining on 6 mm sieve, passing #40 sieve, and remaining on #40 sieve and are shown in Table 5-3. Overall means for these three fractions were 296 ng/g, 253 ng/g and 486 ng/g for that remaining on a 6 mm, passing the #40 sieve, and remaining on the #40 sieve, respectively. In computing the means, extremely high numbers, i.e., > 10,000 ng/g, were excluded. This amounted to three numbers being excluded from the entire data set. These were 21,600 ng/g for the fraction remaining on 6 mm sieve, 70,000 ng/g and 10,400 ng/g for the fraction remaining on #40 sieve. It was assumed that these extremely high values resulted from sampling directly into a battery or some other Hg containing device and were not representative of the concentrations typically found in SW (see Fig. 5-1).

Table 5-3 shows that the largest concentrations of Hg were found at LFS 3. No data are shown for the fraction passing a #40 sieve at this site because these samples were missing from storage. Mercury concentrations for the other two fractions are about 4 and 7 times those found at LFS 4, the most appropriate site of the five for comparison with LFS 3 (i.e., both being approximately the same age and both being unlined sites). No explanation can be given for this finding. However, it has been suggested (Dr. J. F. K. Earle, Personal Communication) that perhaps this site was the recipient of medical and biological wastes that the landfill routinely accepted at the time LFS 3 was being filled. These wastes may have contained higher concentrations of Hg than conventional MSW. In the discussion that follows, one should keep in mind that something was definitely different about the MSW that was loaded into LFS 3 than that for the other four sites.

In the fraction remaining on the 6 mm sieve, Hg concentrations ranged from 187 ng/g at LFS 4 to 761 ng/g at LFS 3. In the passing #40 fraction the values ranged from 65

Table 5-3 Mercury Means in different fractions at different sites

Fractions	LFS 1	LFS 2	LFS 3	LFS 4	LFS 5
Ret on 6 mm	289	208	761	187	217
Passing #40	133	164	N/A	65	427
Ret on #40	396	440	1085	163	502

All units are in ng/g

ng/g at LFS 4 to 427 ng/g at LFS 5. In the remaining on #40 fraction, the values ranged from 163 ng/g at LFS 4 to 1085 ng/g at LFS 3. In general, lower Hg concentrations were detected at each site in the fraction passing #40 sieve which was expected since that fraction contained mostly sand and had the lowest content of volatile solids (Table 5-4). The lower organic matter contents in these fractions probably accounted for the lower amounts of Hg that were bound to these residues.

At all sites, except LFS 4, the highest Hg concentrations were in the fraction remaining on the #40 sieve. Since the fraction remaining on the 6 mm sieve generally had a higher volatile solids content than that remaining on a #40 sieve (Table 5-4), this suggests that the organic matter in the latter fraction was more reactive toward Hg. This could have been due to a larger surface area and/or a more decomposed state for organic matter in the fraction passing the 6 mm sieve compared to that remaining on the 6 mm sieve.

From Table 4-1, the usual trend noted in the fractions at the sites was increasing Hg with increasing depth, except at LFS 1 which had leachate recycling. Leachate recycling is believed responsible for distributing Hg throughout the depths, so that there was no concentration effect at the

Table 5-4 MSW Sample Volatile Solids Content Average by Round and Area

	% VS Above 6 mm	% VS Above #40	%VS Pass #40
Round 3			
LFS1	72.4%	53.7%	5.5%
LFS2	81.3%	50.9%	4.1%
LFS1*	69.9%	48.4%	6.2%
Round 4			
LFS1	80.7%	41.0%	7.0%
LFS1*	77.4%	38.8%	6.6%
LFS5	87.2%	64.4%	5.6%
Round 6			
LFS1	89.2%	58.1%	6.2%
LFS2	90.2%	59.6%	9.3%
LFS1*	89.5%	49.4%	5.8%
Round 7			
LFS2	----	45.9%	2.5%
LFS5	----	57.6%	4.1%
Round 8			
LFS1	----	41.3%	3.3%
LFS2	----	54.6%	3.7%
LFS1*	----	34.9%	3.4%
LFS4	----	41.6%	10.3
Round 9			
LFS1	64.7%	36.7%	3.5%
LFS2	78.5%	54.4%	4.6%
LFS1*	64.7%	35.3%	4.0%
LFS3	77.0%	31.1%	4.8%
LFS5	77.7%	61.4%	4.4%

*Combined fraction of material above 6 mm screen and material Above the #40 sieve
(Miller *et al.*, 1986)

lowest depth The sites without leachate recycling indicated that there was possibly some leaching of Hg taking place If that is the case, then for unlined landfills, there is a possibility that Hg could be getting into the groundwater system Christensen et al (1994) reported Hg concentrations in landfill leachate for landfills of less than 20 years of age These concentrations ranged from 0 05 to 160 µg/L Thus, the potential for Hg to leach through landfills is quite variable but could certainly occur with Hg concentrations at the upper end of this range

The volatilization of Hg from the Alachua County Landfill to the atmosphere was not evaluated during Phase 1 of this study However the potential for Hg volatilization during anaerobic digestion of MSW was evaluated during Phase II

Phase II

An important part of the Phase II experiments was to determine Hg levels in the gaseous phase Quite a few authors had indicated that impregnated activated carbon was capable of absorbing elemental Hg (Vidic and McLaughlin, 1996 Krishnan et al 1994; and Moffitt and Kupel 1971) It was stated that sulfur impregnated activated carbon enhanced Hg removal over those non-impregnated types due to HgS formation on the carbon surfaces. This chemisorption process was enhanced by temperatures between 25°C and 90°C, resulting in increased removal of elemental Hg. It was stated that at a temperature of 140°C, there was a decrease in adsorptive capacity, thus a decrease in HgS formation (Vidic and McLaughlin, 1996; Krishnan et al., 1994). The problem with these reports was that they did not indicate how to desorb the trapped Hg from the impregnated activated carbons. Desorbing trapped Hg was imperative in this study in order to quantify the amount of Hg volatilized during anaerobic digestion.

Krishnan et al. (1994) used high temperatures to desorb the trapped Hg from the carbon while Moffitt and Kupel (1971) stated that the impregnated charcoal was analyzed by AAS. The S-impregnated activated carbon used in the method development and experiments in Phase II was obtained from Norit Americas and they state that it is capable of trapping 99.9% of the elemental Hg in a gas stream. This proved to be true in the initial experiments of this study that were performed to determine trapping efficiency. It did trap all of the elemental Hg that was passed through it, but the problem that arose was how to desorb the Hg from it. The USEPA Method 7471 for determination of total Hg in solid material was not sufficient to enable desorption of the trapped Hg. Therefore, a modification of this method was developed to recover the trapped Hg from the carbon. This modification is shown in Figure 3-10. The development of this method was very important for the Phase II experiments because it was important to quantify the Hg that existed in the gaseous phase. The modified method yielded >98% recovery of the Hg that was sorbed to the S-impregnated activated carbon.

Additional experiments that were performed involved the setup of a system to test two hypotheses: (1) that tygon tubing may be able to trap elemental Hg and Hg^{2+} at 50°C, and (2) that Hg^{2+} in the nitrate form may be volatilized at 50°C and trapped on the S-impregnated activated carbon. The Merck Index (1989) states that $HgCl_2$ volatilizes unchanged at 300°C and volatilizes appreciably at 100°C. Thus it was important to know if Hg^{2+} in the nitrate form could volatilize at 50°C. This was a 60 hour experiment that was set up in an incubator room that operated at 50°C. There were four flasks altogether, hooked up to S-impregnated activated carbon traps and also attached to a pump that had a total flow rate of 30 mL/min. All four flasks contained DDDI water and were spiked with 100 mg Hg^{2+} as $Hg(NO_3)_2$. However, two flasks also received $SnCl_2$ to reduce the Hg^{2+} to Hg^0.

It was found that essentially all of the Hg in the two flasks that received $SnCl_2$ was trapped on the carbon and there was none in the tygon tubing, thus indicating that even at 50°C, tygon tubing is not able to trap elemental Hg. In the case of the two flasks that did not contain $SnCl_2$, no Hg was found in the tygon tubing, but 30-35% of the Hg was found in the carbon traps. This proved that Hg^{2+} has the ability to volatilize directly at 50°C, and that it is not trapped in the tubing. Thus, any Hg found in the tygon tubing cannot be attributed to volatilization of elemental Hg or Hg^{2+}. The results of this experiment show that if Hg is found in the tygon tubing following the reactor experiments, then other forms of Hg were probably being volatilized. Tygon tubing has the ability to trap organic vapors (Dr. R.D. Rhue, Personal Communication), therefore in the case of Hg, the forms that would be expected to be trapped in the tubing would be the organic forms, possibly including dimethylmercury. Also, the death in 1997 of Dr. Karen Wetterhahn resulted from dimethylmercury penetrating her latex gloves indicating that polymer type materials can absorb dimethylmercury.

Phase II was conducted in order to determine the fate of Hg in landfills and involved the use of anaerobic digesters to simulate the processes occurring in a landfill. These digesters allowed precise control over experimental conditions as well as a mass balance in determining the fate of Hg. The mixture of the SW used in the simulated landfill systems or reactors was assumed to be representative of the normal waste stream after recycling (Owens and Chynoweth, 1993). Paper contributes 30 to 50% of MSW and is normally considered the largest component (US Congress, 1989), while food waste represents roughly 10% of MSW (Tchobanoglous, 1993). The biodegradable portions of MSW would include paper and food waste. In the report by Owens and Chynoweth (1993), biochemical methane potentials (bmp) were determined for yard waste, office paper, printed and unprinted newspaper and food packaging. The ultimate methane yields that were determined from these components provided a basis on which to evaluate the digester results in this study. For office paper, unprinted newspaper, and printed newspaper, the ultimate methane yields were 0.369, 0.084, and 0.100 m^3/kg, respectively. Using these numbers as a basis for ideal behavior in anaerobic digesters, it was important for the methane yields to fall within these parameters. Figures 4-10, 4-11, 4-12, 4-25, 4-26, 4 27, and 4-28 show this to be true.

There are four major microbial steps involved in the anaerobic digestion process, namely 1) Hydrolysis, 2) Fermentation, 3) Acidogenesis, and 4) Methanogenesis. Hydrolysis involves hydrolytic bacteria breaking down complex organic compounds (carbohydrates, lipids, and proteins) into simpler units known as monomers. Fermentation involves fermentative bacteria acting on these monomers to produce organic acids, alcohols, neutral compounds, hydrogen, and carbon dioxide. During acidogenesis, some of the fermentation products are converted to hydrogen, carbon dioxide, and acetate by acid-producing bacteria. Specifically, hydrogen-producing acetogenic bacteria are responsible for converting hydrolysis and fermentation products to acetate, hydrogen, and carbon dioxide, while homoacetogenic bacteria ferment hydrogen, carbon dioxide, and formate to acetate. In the fourth and final stage called methanogenesis, two types of methane producing bacteria are active. One converts acetate to methane and carbon dioxide and the other reduces carbon dioxide to methane (Earle et al, 1991; Chynoweth et al., 1984).

Therefore, to determine whether or not a digester is operating as it should under anaerobic conditions, it must exhibit these steps. The two most notable steps would be the acid-forming step which would be represented by a drop in pH and the methanogenic step which would be represented by a steady increase in methane content until there is a stabilization at around 55%.

The data in Figures 4-1 to 4-28 indicated that the reactors in both runs were behaving as expected under anaerobic conditions. In Run 1, the pH data showed a decrease from originally 7.5 to 6.5 at roughly day 5 indicating that the acid forming bacteria were active at that time. The control, 100 ng, and 2000 ng Hg reactors showed consistent data with a decrease in pH from above neutral to about 6.6. The methane content in the gas phase appeared to peak at 55% at around day 7 and then was maintained for the following days. At 100 ng and 2000 ng Hg, there was some fluctuation in the methane content at the later days.

In Run 1, the VFA peaked at day 5 to about 1600 mg/L for the control reactors and 1100 mg/L for the 100 ng Hg reactors and tapered off for the following days. The 2000 ng Hg reactors peaked to 2400 mg/L at day 5 for Reactor 7 and 1000 and 1400 for Reactors 8 and 9, respectively. Additional peaks were noticeable at other days for unknown reasons. Methane yield graphs (Figures 4-10 to 4-12) were all quite consistent and indicated that the reactors were operating properly. They yielded greater than 0.100 m^3/Kg "VS added" which indicated that they were producing adequate methane yields.

In Run 2, the pH's in all of the reactors appeared to decrease from 7.6 to 6.6 at day 3 to day 4. This implied that the acid-forming bacteria came into play one day earlier compared to Run 1. The percent methane (Figure 4-17 to 4-20) also peaked at about 53% at an earlier day, i.e. day 4. The VFAs in Figure 4-21 increased to 2500 mg/L at day 3 and then to greater than 7000 mg/L at day 7. In Figure 4-22, the VFAs increased to 2000 mg/L at day 3 and then to greater than 6000 mg/L at day 8. In Figure 4-23, there was a marked increase in the VFAs at day 3 to about 2000 mg/L and then increased between days 6 and 8 to greater than 5000 mg/L. In Figure 4-24, the VFAs showed an increase to 1800 mg/L at day 3 and then an increase to greater than 4000 mg/L at day 6. The methane yields for the reactors in Run 2 were consistently higher than those for Run 1. In Run 2, silicone, in conjunction with teflon tape, was used to seal all fittings and connectors, while in Run 1 only teflon tape was used. The silicone along with the teflon tape seemed to make the reactor system more air-tight. The gas production in Run 2 was higher than Run 1 even though both methane yields fell within acceptable limits, i.e., > 0.100 m^3/Kg "VS added".

In Figure 4-27, the methane yield for reactor 7 (R7) was lower than that for reactor 6 (R6). R7 was nearly 0.25 m^3/Kg "VS added" whereas R6 was 0.20 m^3/Kg "VS added". Lab notes showed that air was accidentally let in. Figure 4-28 showed a lower methane yield in R9 than R8. R8 showed a value of 0.23 m^3/Kg "VS added" whereas R9 showed a value of 0.17 m^3/Kg "VS added". Although R9 had a lower value than R8, it was still within the acceptable limits stated previously. R9 had several problems including leaky septum. These problems were not "fatal" to the system, but were considered nuisances.

To help explain the data, it is necessary to understand the role of microorganisms in these systems. During anaerobic digestion, the same microorganisms responsible for producing methane may influence the transformations of Hg in this system. In this system the main transformations which occur are reduction and methylation. Mercury must be in the divalent form for methylation to occur. Mercury can be methylated by anaerobic microorganisms such as those listed above, to form either mono- or dimethylmercury. The transformations are shown below.

$$Hg^{2+} + RCH_3 \longrightarrow CH_3Hg + R \qquad (1)$$

$$2CH_3Hg^+ + H_2S \longrightarrow (CH_3)_2Hg + HgS + 2H^+ \qquad (2)$$

The formation of mono and dimethylmercury depends on the Hg concentration and the pH of the system. Monomethylmercury (1) is formed under acidic pH (low pH) conditions and in the event that Hg concentrations are high. Therefore, the bacteria responsible for producing acetate during methane fermentation would be creating favorable conditions for monomethylmercury production. Dimethylmercury (2) is normally formed under alkaline or neutral conditions when strong complexing agents such as hydrogen sulfide (H_2S) are present or when Hg concentrations are low. As a result, dimethylmercury may be observed in an anaerobic digestion system. The reaction rate constant for synthesis of monomethylmercury is 6000 times larger than that for dimethylmercury. Thus in most environmental systems only about 3% of methylmercury is dimethylmercury (Regnell 1990). In the case of anaerobic digesters (reactors) this percentage may be greater since the system exists under mostly alkaline conditions which are more conducive to formation of dimethylmercury

Lindqvist and Schroeder (1989) implied that transformations of divalent Hg to monomethylmercury by bacteria are mechanisms of detoxification and excretion Gilmour and Henry (1991) suggested that sulfur-reducing bacteria may also produce methylmercury, but due to the fact that a lot of sulfur is not present in an anaerobic digestion system, it could be stated that mostly methanogens and a small amount of sulfur reducers are responsible for methylmercury production The sulfur that is present, especially in the form of hydrogen sulfide would transform some of the divalent Hg to mercuric sulfide (HgS) which is extremely insoluble and would precipitate (Gilmour and Henry, 1991)

The last transformation that can occur is the reduction of monovalent and divalent Hg to elemental Hg which can be lost to the atmosphere Also, some microorganisms contain a detoxification mechanisms for methylmercury that reduces it to elemental Hg plus methane (Brock and Madigan, 1988) Some of the transformations are shown below

$$CH_3\text{-}Hg\text{-}CH_3 \xleftarrow{\text{bacterial}} CH_3Hg \xrightarrow{\text{bacterial}} Hg^0 + CH_4$$

$$^{*}Hg\text{-}Hg \xrightarrow{\text{chemical}} Hg^{2+} \overset{\text{bacterial}}{+ Hg^0}$$

$$Hg^{2+} \xrightarrow{\text{bacterial}} Hg^0$$

Therefore, all of the above transformations of Hg may result from the action of some of the organisms involved in methane fermentation, particularly, methanogens, sulfur-reducing bacteria, and hydrogen producing bacteria.

From this microbial information one can develop an understanding of the Hg data from the anaerobic reactors that was presented in Figures 4-31 to 4-35. It was clear that in all of the cases, i.e., control reactors and the others spiked with 100, 1000, and 2000 ng Hg that the majority of the Hg was held tightly in the SW. With the 100 ng Hg spike, the range of Hg in the gas phase ranged from 30% to 48% of the total. It was previously stated that dimethylmercury can be formed microbially under alkaline conditions and with lower levels of Hg. Therefore the gaseous phase Hg could have been a mixture of dimethylmercury along with elemental Hg. Evidence for this was the fact that approximately 45% of the Hg that was volatilized in this treatment was found in the tygon tubing, which was shown not to react with either Hg^{2+} or Hg^0. There were generally only trace levels of Hg in the leachate indicating that the Hg that had not volatilized was tied up in the SW itself.

In the case of the anaerobic reactor with the 1000 ng spike, Hg in the gas phase accounted for 8.3% to 12.6% of the total added. This was lower than that with the 100 ng spike and this could possibly be attributed to some inhibition of the microbes that were responsible for converting the Hg to a volatile form, although methane, VFA, and total gas production gave no evidence that Hg inhibited the other anaerobic digestion processes. The higher concentration of Hg in the SW on which the microbes fed may have contributed to this reduction of Hg in the gas phase. The low Hg concentration in the leachate and the fact that 37% of the Hg volatilized was found in the tygon tubing also implicate dimethylmercury as one form of Hg that was volatilized since dimethylmercury is formed at low levels of Hg. Another possibility could be that at least some of the Hg found in the gaseous phase was elemental Hg which had been converted directly by certain microbes from methylmercury and divalent Hg.

In the case of the anaerobic reactor with the 2000 ng Hg spike, the Hg in the gas phase ranged from 2.1% to 6.6%, but most of the reactors were in the 2.1% to 2.5%, with only one indicating 6.6%. Figure 5-3 shows the different Hg spiking levels in ng and the Hg volatilized (ng) for both runs. This figure is not based on the percent volatilized, but on the actual mass of Hg (ng) that was volatilized. The data appear to be quite variable, especially for the 1000 and 2000 ng Hg treatments. For the 100 ng Hg spike, the Hg volatilized ranged from 30 to 48 ng Hg. For the 1000 ng Hg spike, the Hg volatilized ranged from about 85 to 120 ng Hg, whereas in the 2000 ng Hg spike, the Hg volatilized ranged from 42 to 136 ng Hg. This figure shows the variability in the amounts of Hg volatilized is both runs. It is possible that this same variability will be observed when sampling the gas phase of landfills to determine Hg volatilization.

Key microbes in the conversion of divalent Hg to elemental Hg and dimethylmercury or methylmercury to elemental Hg and methane should exist in these reactors and may have been inhibited or out competed by other microbes in the system as Hg concentrations increased. *Escherichia coli* is one

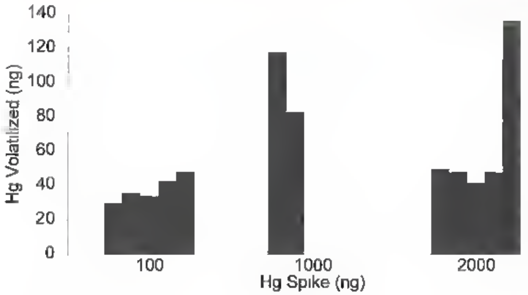

Figure 5-3 Hg spike (ng) in simulated landfill reactors & amount of Hg volatilized (ng)

microbe that can absorb divalent Hg into its system where it combines with the cytoplasm and is converted into a different compound or the volatile elemental form. Other microbes adsorb divalent Hg on the outside of their cell walls where it is converted directly into a volatile form (D'itri and D'itri, 1977). It has also been stated that the enzymes involved in Hg volatilization are chromosomally encoded in specific bacterial genera, namely *Staphylococcus aureus* and *Bacillus spp.* (Nakamura et al., 1990).

The chemistry involved in the Hg binding and precipitation taking place in these anaerobic reactors is complex. In anaerobic environments there is a tendency for Hg to be precipitated as the sulfide (HgS). The insolubility of HgS makes it resistant to methylation, but under aerobic conditions, HgS may be oxidized to the sulfate form which can then undergo methylation. Under low Eh and high pH conditions, HgS can be transformed to HgS_2^{2-}, or to the free metal (Barkay, 1992; Wilken, 1992, Schuster, 1991). The binding of Hg in SW may occur in a similar fashion as it would in soil. Some of these binding mechanisms would include: (1) specific adsorption - strong binding due to covalent or coordinated forces; (2) chelation - bound to organic substances, and (3) precipitation - as sulfide, carbonate, hydroxide, phosphate, etc. The two most probable mechanisms would probably be chelation due to the high organic matter in SW and precipitation, especially in a reduced environment with sulfide present. An interesting finding from this study is that there is evidence that landfills could be responsible for Hg emissions into the atmosphere. This has not been shown before. Other sources such as municipal waste incinerators and medical waste incinerators are thought to be more important sources of Hg going into the atmosphere as result of waste disposal. Using the data obtained from this study, an estimate of the amount of Hg emitted from landfills in the state of Florida was made. This calculation was made using the following assumptions: 1) the amount of MSW going to landfills per person per day is 8 pounds, 2) the population of Florida is 15 million, 3) the average Hg concentration in MSW going into the landfills is 200 ng/g, and 4) the percentage of the Hg that volatilizes from MSW is 30%. Using these assumptions, the annual amount of Hg volatilizing from landfills in the state of Florida is about 2600 pounds. To put this number into perspective, Chu and Porcella (1995) reported that annual Hg emissions from total electric utilities in the United States was 40 tons. If these estimates are correct, then Hg emissions from landfills to the atmosphere could account for a considerable fraction of anthropogenic Hg sources. This finding may be particularly important with regard to South Florida where the problem of atmospheric Hg pollution has been well documented (Reed et al., 1995).

CHAPTER 6
SUMMARY AND CONCLUSIONS

This study has provided important information about the fate of mercury in municipal landfills. Phase I of the landfill experiments indicated that mercury does exist in the Alachua County landfill, and by implications, other landfills as well. The investigations in Phase II followed the Phase I work and showed that landfills could be a potential source of mercury to the atmosphere. The conclusions drawn from the two phases of this study are summarized below.

Phase I

1. Over half of the samples analyzed for Hg had concentrations below 150 ng/g. However, the distribution of Hg was highly skewed with some samples containing thousands of ng Hg/g on a dry weight basis. The highest Hg concentration exceeded 16,000 ng/g. The distribution of Hg in the Alachua County Landfill was extremely heterogeneous. These "hot spots" probably resulted from

samples being taken directly from areas where Hg-containing devices, such as batteries and switches, had decomposed

2 Mercury was found in all samples taken from the Alachua County Landfill While Hg concentrations were all well above background levels for surface soils in Florida they were two to three orders of magnitude lower than either the current soil clean-up goals used by FDEP or maximum allowable concentrations in sewage sludge as mandated in 40 CFR Part 503. Compost samples from Palm Beach County consisting of a 1:1 mixture of yard wastes and sewage sludge were higher in Hg than the landfill samples but still far below the regulatory levels used by FDEP and EPA Therefore, Hg should not be a problem is landfills are reclaimed and the SW residue applied to land

3 Leachate recycling at the Alachua County Landfill resulted in a more uniform distribution of Hg with depth compared to three other locations within the landfill in which leachate was not recycled.

Phase II

1 The bulk of the Hg added to the anaerobic digesters was

found in the solid waste at the end of the experiments In 16 out of 18 leachate samples no Hg was detected Very low levels of Hg in leachate is consistent with landfill leachate data reported in the literature

2 Mercury volatilized during anaerobic digestion of artificial MSW. The percentage volatilized ranged from over 30% at the lowest Hg concentration to about 3% at the highest Hg concentration used The presence of Hg in the tygon tubing connecting the reactor to the carbon traps was taken as evidence that forms of Hg other than Hg^0 and Hg^{2} were volatilized A review of the literature provided strong evidence to support the hypothesis that DMHg can be volatilized under conditions that exist during anaerobic digestion of MSW DMHg and possibly other volatile organic forms of Hg are believed responsible for the Hg associated with the tygon tubing

3. Sulfur impregnated carbon was used to successfully trap Hg that was volatilized during the anaerobic digestion experiments EPA Method 7471 was unable to recover any Hg from these carbon traps However, a modification of the method was successful in recovering >98% of the trapped

Hg, allowing quantitative recovery for the mass-balance calculations

4. The distribution of Hg in the solid waste residue was extremely heterogeneous An accurate determination of the Hg associated with the solid fraction required that the entire sample be analyzed This condition may have resulted from the manner in which the Hg was added to the artificial MSW at the start of the experiment and to the fact that the solid waste at the end of the experiment was not dried and homogenized prior to analysis in order to avoid potential Hg losses during the drying step
5. The amounts of Hg volatilized were highly variable, particularly at the two higher Hg concentrations At the 2000 ng Hg treatment, the amounts volatilized ranged from 42 to 136 ng
6. Run 1 lasted 37 days, Run 2 lasted 16 days. However, the volatilization of Hg was essentially the same for both runs, indicating that volatilization may occur only during the early stages of digestion

From these conclusions, certain recommendations can be made These are as follows:

(1) The recycling of mercury containing devices and lamps must increase if the amount of mercury that is making its way into landfills is to decrease further

(2) Landfill gases should be monitored for mercury and if Hg is being volatilized, special traps should be placed on landfills to capture mercury that may be escaping into the atmosphere

(3) The effect of leachate recycling on the distribution of Hg in landfills should be evaluated further If the results observed in Phase I of this study hold true for other landfills, then this effect should be factored in when determining the value of leachate recycling at new installations

(4) Further studies should be performed on landfills especially in the South Florida region, to determine the contribution of mercury from landfills to the atmosphere This would allow for a better understanding of the mercury problem that exists in the Florida Everglades and surrounding systems

(5) Additional research should be designed to determine at what stages during anaerobic digestion of MSW Hg is being volatilized It should also determine whether there are

stages during anaerobic digestion when Hg can be found in the leachate. Hg concentrations from the leachate recycling area of the Alachua County Landfill suggest that there is a stage during anaerobic digestion when Hg is mobile and redistribution with depth in a landfill can occur.

APPENDIX A
RAW MERCURY DATA FOR ALACHUA COUNTY LANDFILL RESIDUES AND PALM BEACH COUNTY COMPOST

Table A 1. Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I D	Absorb	Hg	Sample Wt	[Hg]	Comments
		(000 0nm)	(ng)	(g)	(ng/g)	(% rec y)
00/00/00	Blank	0 011	0			
	Std 1 (00 ng)	0 010	00			
	Std 0 (000 ng)	0 005	54			
	Std 0 (000 ng)	0 000	157			
	Std 0 (200 ng)	0 100	000			
	Std 0 (200 ng)	0 020	200			
	Std 0 (100 ng)	0 000	100			
	Std 7 (000 ng)	0 060	340			
	Std 0 (400 ng)	0 000	301			
	Std 5 (400 ng)	0 200	040			
	Std 10 (000 ng)	0 012	500			
	Check Std	0 071	007			
	LFS 1 0100 (25 00) pass H0	0 070	010	0 0000	000	
	LFS 0 0100 (25 00) pan on H0	0 000	000	0 0000	300	
	LFS 1 0100 (00 00) 0	0 000	000	0 0000	000	
	LFS 0 0100 (30 00) pass H0	0 000	000	0 0000	003	
	LFS 0 0002 (30-40) pass H0 (DUP)	0 000	007	0 0000	017	
	LFS 0 0000 (00 00) ren on 000	0 000	000	0 0020	007	
	LFS 0 0002 (0 00) C	0 000	70	1 0000	01	
	LFS 0 0000 (00 20) C	0 000	000	0 0000	100	
	LFS 0 0000 (00 00) C pass 000	0 000	70	1 0207	70	
	LFS 0 0000 (00 00) C pass 000 (00)	0 000	000	0 0211	012	00 942045
	LFS 0 0000 (15 00) C ren on 000	0 000	007	1 8101	327	
	LFS 0 0000 (25 00) C ren on 000	0 000	000	1 1020	000	
	CCB	0 017	0			
	CCV (00 ng Hg)	0 000	00			
	LFS 0 0000 (5 15) pass 0 40	0 000	00	0 0001	00	
	LFS 0 0000 (0 15) ren on 0 00	0 100	185	0 0000	150	
	LFS 0 0000 (25 00) pass 0 00	0 000	87	0 0200	00	
	LFS 0 0000 (00 00) ren on 0 00	0 100	101	0 0000	000	
	LFS 0 0701 (0 10) pass 0 40	0 002	6	0 0200	0	
	LFS 0 0700 (0 10) pass 0 00 (DUP)	0 000	10	0 0000	00	
	LFS 0 0701 (0 00) ren on 0/0" L 0 0	0 001	51	0 0000	00	

Table A 1 (con't d)   Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I D	Absorb	Hg	Sample Wt	[Hg]	Comments
12/10/05		(253 6nm)	(ng)	(g)	(ng/g)	(# cac y)
	LFS 2 0701 (10 20) pass #60	0 020	20	1 0071	20	
	LFS 2 0701 (10 20) ret on 1/4" & #4	0 098	115	1 0138	113	
	LFS 2 0701 (20 30) ret on 1/4" & #4	0 170	175	1 0107	173	
	LFS 2 0702 (25 27 5") pass #40	0 242	520	1 0175	511	
	CCB	0 010	0			
	CCV (50 ng Hg)	0 040	40			

Table A.[illegible] (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample ID	Absorb ([illegible] nm)	Hg (ng)	Sample Wt	[Hg] (ng/g)	Comments ([illegible])
[illegible]/20/[illegible]	Blank	[illegible]	[illegible]			
	Std [illegible] ([illegible] ng)	[illegible]	53			
	Std [illegible] ([illegible] ng)	[illegible]	150			
	Std [illegible] (25[illegible] ng)	[illegible]	[illegible]			
	Std [illegible] (3[illegible] ng)	[illegible]	[illegible]			
	Std [illegible] ([illegible] ng)	[illegible]	[illegible]			
	Std [illegible] ([illegible] ng)	[illegible]	[illegible]			
	Check Std	[illegible]	[illegible]			
	LFS [illegible] ([illegible])	[illegible]	[illegible]	[illegible]	[illegible]	Redo
	LFS [illegible] ([illegible])	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible])	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) pass [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	Redo
	LFS [illegible] ([illegible]) pass [illegible] (DUP)	[illegible]	[illegible]	[illegible]	[illegible]	Redo
	LFS [illegible] ([illegible]) rem on [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	Redo
	LFS [illegible] ([illegible]) pass [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) pass [illegible] (SPIKE)	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	LFS [illegible] ([illegible]) rem on [illegible] & [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) pass [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) rem on [illegible] & [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) [illegible] rem on old	[illegible]	[illegible]	[illegible]	[illegible]	
	CCB	[illegible]	[illegible]			
	CCV ([illegible] ng Hg)	[illegible]	[illegible]			
	LFS [illegible] ([illegible]) C3rd pass [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) C3rd pass [illegible] (DUP)	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) C3rd rem on old	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) A 3rd pass [illegible]	0.045	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) A 3rd rem on [illegible]	0.074	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) A 3rd rem on old	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) [illegible] biodegrad (mixed)	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) [illegible] biodegrad (mixed)	[illegible]	[illegible]	[illegible]	[illegible]	
	LFS [illegible] ([illegible]) C3rd rem on [illegible]	[illegible]	[illegible]	[illegible]	[illegible]	

Table A 1 (con t d)   Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I D	Absorb (253 7nm)	Hg (ng)	Sample Wt	[Hg] (ng/g)	Comments (% recly)
11/10/00	LFS 2 0001 (15 10) A3rd rem, as old	0 020	0	2 0110	0	
	LFS 1 0001 (10 10) C3rd pass #40 (SP)	0 071	111	2 0170	110	
	CCB	0 015	0			
	CCV (00 ng Hg)	0 030	00			
	LFS 1 0001 (0 10) pass #40 (old)	0 101	101	1 0100	105	
	LFS 1 0001 (10 10) pass #00 (A)	0 000	1001	1 0111	1007	Redo
	LFS 1 0001 (0 10) oll #10 fine ground	0 010	131	1 0110	111	
	LFS 2 0001 (10 10) oll #10 fine ground	0 110	190	1 0130	100	
	LFS 1 0101 (10 10) pass 000	0 110	030	1 0121	111	
	LFS 2 0001 (10 10) pass 000 DUP)	0 112	221	2 0151	110	
	LFS 1 0001 (10 00) (old) final ground sampl	0 140	110	2 0271	110	
	LFS 1 0001 (10 10) (new) final ground sampl	0 111	101	2 0027	110	
	LFS 0 0001 (10 10) all bis ground fine	0 010	111	1 0100	131	
	LFS 0 0001 (10 25) all bis ground fine	0 111	171	2 0120	301	
	LFS 1 0001 (10 10) (old) final sample	0 111	015	1 0100	001	
	LFS 1 0001 (0 10) res org on 1/0 screen	0 111	115	1 0040	101	
	LFS 1 0001 (11 15) res org on 1/0 screen	1 015	1001	1 0301	1011	Redo
	LFS 1 0001 (1 11) res org on 1/0 screen	0 126	111	1 0125	111	
	LFS 1 0001 (11 11) res org on 1/0" screen	0 101	110	1 0107	100	
	CCB	0 017	1			
	CCV (50 ng Hg)	0 110	01			
	LFS 1 0001 (1 15) res org on 1/0" screen	0 110	116	1 0010	117	
	LFS 1 0001 (15 11) res org on 1/0" screen	0 091	111	1 0101	170	
	LFS 1 0001 (21 11) res org on 1/0" screen	0 110	111	1 0100	100	
	LFS 1 0101 (10 10) res org on 1/0" screen	0 130	111	1 0110	105	
	LFS 1 0101 (20 10) res org on 1/0" screen	0 110	101	1 0100	001	
	LFS 1 0101 (0 10) res org on 1/0" screen	0 011	00	1 0110	00	
	LFS 1 0001 (20 30) res org on 1/0" screen	0 001	110	1 0112	100	
	CCB	0 010	0			
	CCV (00 ng Hg)	0 011	10			

Table A 1 (cont'd). Raw Mercury Data for Alachua County Landfill Residues and Palm Beach County compost

Date	Sample I.D	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)	Comments (% rec'y)
1/12/98	Blank	0.018	0					
	Std 1 (50 ng)	0.034	43					
	Std 2 (150 ng)	0.098	150					
	Std 3 (250 ng)	0.138	250					
	Std 4 (350 ng)	0.180	356					
	Std 5 (450 ng)	0.228	457					
	Std 6 (600 ng)	0.265	538					
	Check Std	0.078	124					
	LFS 5 0401 (20-30')	0.111	201	1.0083	199	20/80 dilution	781	
	LFS 5 0401 (30-40') pass. #40	0.050	67	1.0211	66	1/50 dilution	3303	
	LFS 5 0402 (30-40') rem on #40	0.060	166	1.0085	160	1/65 dilution	10384	
	LFS 1 0801 (10-20') (A) pass #40	0.028	8	1.0152	8	1/50 dilution	413	
	LFS 1 0801 (10-20') (A) pass #40 (DUP)	0.022	6	1.0152	6	1/50 dilution	306	
	LFS 1 0801 (10-20') (A) pass #40 (TRIPLICATE)	0.023	8	1.0160	8	1/50 dilution	413	
	LFS 1 0901 (25-35') rem org on 1/4" screen	0.145	274	1.0210	270	1/80 dilution	21590	
	PALM BEACH COUNTY COMPOST							
	301	0.258	522	1.0110	517			
	302	0.312	641	1.0178	630	Redo		
	303	0.304	623	1.0110	616	Redo		
	303 (DUP)	0.520	1098	1.0139	1084	Redo		
	304	0.350	724	1.0161	712	Redo		
	305	0.104	186	1.0169	183	20/80 dilution	733	
	306	0.125	232	1.0160	228	20/80 dilution	914	
	307	0.127	236	1.0142	233	20/80 dilution	930	
	307 (SP)	0.147	280	1.0145	276	20/80 dilution	1103	87.5
	308	0.093	162	1.0181	159	20/50 dilution	396	
	309	0.068	102	1.0112	101	20/80 dilution	406	
	310	0.048	107	1.0177	105	20/80 dilution	420	
	CCB	0.018	0					
	CCV (50 ng Hg)	0.036	45					

Table A 1 (cont'd) Raw Mercury Data for Palm Beach County Compost

Date	Sample I.D.	Absorb. (253.7 nm)	Hg (ng)	Sample Wt.	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
1/18/06	Blank	0.010	0				
	Std 1 (50 ng)	0.039	45				
	Std 2 (150 ng)	0.088	152				
	Std 3 (250 ng)	0.132	250				
	Std 4 (350 ng)	0.164	335				
	Std 5 (450 ng)	0.212	446				
	Std 6 (550 ng)	0.268	567				
	Check Std	0.073	124				
	302	0.124	242	1.0187	237	20/50 dilution	590
	302	0.117	226	1.0106	223	20/50 dilution	558
	304	0.122	237	1.0196	233	20/50 dilution	581
	311	0.050	94	1.0080	93	20/80 dilution	372
	311 (DUP)	0.053	101	1.0085	100	20/80 dilution	398
	312	0.057	110	1.0116	109	20/80 dilution	436
	313	0.603	1351	1.0156	1330	Redo	
	314	0.281	529	1.0112	523	Redo	
	314 (SP)					Redo	
	315	0.111	212	1.0110	210	11/66 dilution	1250
	CCB	0.010	0				
	CCV 150 ng Hg	0.039	45				
	316	0.142	284	1.0106	281	11/66 dilution	1686
	317	0.086	154	1.0192	151	20/50 dilution	378
	318	0.050	94	1.0198	92	20/80 dilution	368
	319	0.060	116	1.0192	112	20/80 dilution	450
	320	0.084	149	1.0114	148	20/80 dilution	591
	321	0.138	287	1.0143	284	11/66 dilution	1698
	322	0.222	468	1.0142	462	11/66 dilution	2773
	323	0.144	288	1.0160	284	11/66 dilution	1702
	324	0.137	272	1.0164	268	11/66 dilution	1608

Table A-1 (con't'd) Raw Mercury Data for Palm Beach County Compost

Date	Sample I.D.	Absorb. (253.7 nm)	Hg (ng)	Sample Wt.	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
	325	0.067	110	1.0100	108	20/55 dilution	433
	325 (DUP)	0.065	108	1.0132	104	20/54 dilution	418
	CCB	0.019	8				
	CCV	0.038	43				

Table A.1 (con't 6) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% recv'y)
3/14/96	Blank	0.015	0			
	Std 1 (50 ng)	0.037	43			
	Std 2 (100 ng)	0.061	152			
	Std 3 (250 ng)	0.138	269			
	Std 4 (350 ng)	0.188	348			
	Std 5 (450 ng)	0.188	438			
	Std 6 (500 ng)	0.242	552			
	Check Std	0.072	130	1.0069	129	
	LFS 1 0301 (5-15') (A)	0.048	70	1.0177	69	
	LFS 1 0301 (5-15') (A) pass. #40	0.030	46	1.0187	45	
	LFS 1 0301 (5-15') (A) rem. on #40	0.090	195	1.0108	193	
	LFS 1 0301 (15-25') (A)	0.096	164	1.0137	162	
	LFS 1 0301 (15-25') (A) pass #40	0.031	78	1.0299	76	
	LFS 1 0301 (15-25') (A) pass #40 (DUP)	0.048	70	1.0254	68	
	LFS 1 0301 (15-25') (A) rem. on #40	0.103	297	1.0122	294	
	LFS 1 0301 (25-45')	0.079	147	1.0160	145	
	LFS 1 0301 (25-45') pass #40	0.197	415	1.0132	410	
	LFS 1 0301 (25-45') pass #40 (SP)	0.205	460	1.0138	454	99.3
	LFS 1 0301 (25-45') rem. on #40	0.170	373	1.0130	368	
	LFS 1 0302 (5-15')	0.185	410	1.0070	407	
	CCB	0.017	0			
	CCV (50 ng Hg)	0.030	48			
	LFS 1 0302 (5-15') (B) pass #40	0.027	18	1.0179	18	
	LFS 1 0302 (5-15') (B) rem. on #40	0.051	76	1.0205	75	
	LFS 1 0302 (15-25') (B)	0.065	63	1.0096	62	
	LFS 1 0302 (15-25') (B) pass. #40	0.066	80	1.0190	79	
	LFS 1 0303 (5-15') (C) pass #40	0.038	46	1.0157	45	
	LFS 1 0303 (5-15') (C) rem. on #40	0.070	125	1.0130	123	
	LFS 1 0303 (15-25') (C) rem. on #40	0.131	278	1.0168	273	

Table A.1 (con't) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
3/14/96	LFS 1.0303 (25-30') (C)	0.036	41	1.0122	40
	LFS 1.0303 (25-30') pass. #40	0.047	58	1.0168	57
	LFS 1.0303 (25-30') rem. on #80	0.081	103	1.0154	101
	CCB	0.010	0		
	CCV (50 ng Hg)	0.036	45		
	LFS 1.0401 Pond (5-15') organics & rest	0.081	102	1.0124	101
	LFS 1.0401 Pond (25-35') organics & rest	0.040	51	1.0147	50
	LFS 1.0401 Pond (35-45') organics & rest	0.087	111	1.0104	110
	LFS 1.0401 Pond (35-45') pass. #40	0.058	98	1.0085	97
	LFS 1.0401 Pond (35-45') pass. #40 (DUP)	0.082	108	1.0093	107
	LFS 1.0401 Pond (35-45') rem. on #40	0.134	208	1.0098	206
	LFS 1.0402 (15-20')	0.080	98	1.0118	97
	LFS 1.05 pass #40	0.080	100	1.0147	99
	CCB	0.017	0		
	CCV (50 ng Hg)	0.037	43		
	LFS 1.0602 (0-15') all bio mixed	0.220	487	1.0130	481
	LFS 1.0602 (0-15') A3rd rem. on #80	0.280	646	1.0100	640
	LFS 1.0602 (0-15') A3rd rem. on old	0.227	514	1.0109	509
	LFS 1.0602 (15-30') all bio mixed	0.050	88	1.0113	87
	LFS 1.0602 C3rd rem. on old	0.051	78	1.0096	77
	LFS 1.0602 C3rd pass #40	0.060	109	1.0054	108
	LFS 1.0602 C3rd rem. on #40	0.081	152	1.0146	150
	LFS 1.0602 C3rd pass #1 on #40	0.134	258	1.0101	255
	LFS 1.0602 (15-30') C3rd rem. on #80	0.117	242	1.0093	239
	LFS 1.0602 (15-30') C3rd rem. on old	0.054	110	1.0146	108
	CCB	0.018	0		
	CCV (50 ng Hg)	0.036	46		
	LFS 1.0801 (0-10') final (new)	0.120	271	1.0101	268
	LFS 1.0801 (0-10') pass #40	0.154	303	1.0167	298

Table A.1 (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample ID	Absorb. (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
3/14/96	LFS 1 0801 (0-10') pass. #40 (SP)	0.186	413	1.0127	408
	LFS 1 0801 (0-10') rem. on #40	0.144	308	1.0047	307
	LFS 1 0801 (10-20') all bio. ground fine	1.127	2772	1.0018	2767
	LFS 1 0801 (10-20') rem. org. 1/4" & #40	1.074	2619	1.0105	2596
	LFS 1 0801 (20-30') all bio. ground fine	0.141	276	1.0075	274
	LFS 1 0801 (20-30') pass #40	0.129	271	1.0150	267
	LFS 1 0801 (20-30') pass #40 (DUP)	0.130	274	1.0187	269
	LFS 1 0801 (20-30') pass #40	0.130	281	1.0112	278
	CCB	0.015	0		
	CCV (50 ng Hg)	0.036	48		
	LFS 1 0802 (0-10') all bio. fine ground	0.106	219	1.0183	215
	LFS 1 0802 (0-10') pass. #40	0.085	88	1.0116	87
	LFS 1 0802 (0-10') pass #40	0.050	75	1.0067	75
	LFS 1 0802 (0-10') rem. org. on 1/4" & #40	0.100	191	1.0048	190
	LFS 1 0802 (10-20') all bio. fine ground	0.104	205	1.0061	204
	LFS 1 0802 (10-20') pass #40	0.045	63	1.0062	63
	LFS 1 0802 (10-20') pass #40 (DUP)	0.042	56	1.0067	55
	LFS 1 0802 (10-20') pass #40	0.048	70	1.0176	69
	LFS 1 0802 (10-20') rem. org. 1/4" & #40	0.132	277	1.0100	274
	LFS 1 0802 (20-30') all bio. fine ground	0.143	306	1.0066	304
	LFS 1 0802 (20-30') pass #40	0.078	145	1.0092	143
	LFS 1 0802 (20-30') pass #40	0.088	170	1.0087	168
	LFS 1 0802 (20-30') rem. org. 1/4" & #40	0.109	226	1.0087	224
	LFS 1 0901 (15-25') rem. org. on 1/4" screen	0.242	552	1.0165	543
	LFS 1 0901 (35-45') rem. org. on 1/4" screen	0.371	872	1.0199	855
	LFS 1 0902 (25-35') rem. org. on 1/4" screen	0.103	212	1.0125	209
	LFS 1 0902 (35-45') rem. org. on 1/4" screen	0.146	313	1.0002	313
	CCB	0.010	0		
	CCV (50 ng Hg)	0.037	43		

Table A-1 (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)
3/17/98	Blank	0.018	1			
	Std 1 (50 ng)	0.041	51			
	Std 2 (150 ng)	0.077	133			
	Std 3 (250 ng)	0.135	274			
	Std 4 (350 ng)	0.171	347			
	Std 5 (450 ng)	0.210	436			
	Std 6 (550 ng)	0.263	567			
	Check Std.	0.075	131	1.0165	129	
	LFS 2 0301 (0-10')	0.024	13	1.0075	13	
	LFS 2 0301 (10-20')	0.224	488	1.0166	481	
	LFS 2 0301 (10-20') D pass. #40	0.265	607	1.0097	601	Redo
	LFS 2 0301 (10-20') D pass. #40 (DUP)	0.291	620	1.0106	614	Redo
	LFS 2 0301 (10-20') D rem. on #40	1.034	2311	1.0106	2286	
	LFS 2 0301 (20-30') D pass #40	0.204	422	1.0142	416	
	LFS 2 0301 (20-30') D rem. on #40	0.257	543	1.0084	539	
	LFS 2 0302 (0-10') E	0.024	13	1.0082	13	
	LFS 2 0302 (0-10') E pass. # 40	0.034	35	1.0091	35	
	LFS 2 0302 (0-10') E pass. # 40 (SP)	0.058	83	1.0099	80	66.6
	LFS 2 0302 (0-10') E rem. on #40	0.050	72	1.0066	71	
	LFS 2 0302 (10-20') E	0.088	154	1.0084	153	
	LFS 2 0302 (10-20') E pass # 40	0.038	31	1.0127	30	
	LFS 2 0302 (10-20') E rem. on # 40	0.086	151	1.0132	149	
	CCB	0.018	0			
	CCV (50 ng Hg)	0.040	48			
	LFS 2 0302 (20-30') E	0.067	111	1.0071	110	
	LFS 2 0302 (20-30') E pass #40	0.060	98	1.0129	96	
	LFS 2 0302 (20-30') E rem. on #40	0.134	274	1.0122	271	
	LFS 2 0301 (20-30') D	0.098	183	1.0181	181	
	LFS 2 0566 (0-10') all bla. mixed	0.034	35	1.0058	35	

Table A.1 (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)
3/17/99	LFS 2 0603 (15-30') all bio mixed	0.086	149	1.0054	148	
	LFS 2 0603 (15-30') rem. on #40	0.100	166	1.0030	165	
	LFS 2 0603 C3rd (15-30') pass #40	0.072	122	1.0128	120	
	LFS 2 0603 C3rd (15-30') pass #40 (DUP)	0.070	117	1.0113	116	
	LFS 2 0603 C3rd (15-30') rem. on old	0.048	95	1.0043	94	
	LFS 2 0601 (0-10') all bio. fine ground	0.066	109	1.0024	109	
	LFS 2 0601 (0-10') pass. #40	0.048	87	1.0062	87	
	LFS 2 0601 (0-10') rem. org. on 1/4" & #40	0.084	149	1.0011	149	
	LFS 2 0601 (10-20') pass #40	0.048	80	1.0034	80	
	LFS 2 0601 (10-20') rem. org. on 1/4" & #40	0.085	140	1.0094	139	
	LFS 2 0601 (20-30') all bio. fine ground	0.281	598	1.0112	591	Redo
	LFS 2 0601 (20-30') pass #40	0.100	206	1.0058	205	
	LFS 2 0601 (20-30') rem. org. on 1/4" & #40	0.181	370	1.0097	366	
	CCB	0.018	6			
	CCV (50 ng Hg)	0.045	40			
	LFS 2 0601 (30-40') pass #40	0.100	206	1.0055	205	
	LFS 2 0601 (30-40') rem. on 1/4" & #40	0.150	308	1.0064	306	
	LFS 2 0601 (40-45') all bio. fine ground	0.151	313	1.0043	312	
	LFS 2 0601 (40-45') pass #40	0.046	70	1.0120	69	
	LFS 2 0601 (40-45') pass #40 (DUP)	0.050	72	1.0100	71	
	LFS 2 0601 (40-45') rem. on 1/4" & #40	0.080	161	1.0071	160	
	LFS 2 0601 (10-20') all bio. fine ground	0.066	91	1.0088	90	
	LFS 2 0602 (0-10') all bio. fine ground	0.048	67	1.0002	67	
	LFS 2 0602 (0-10') pass #40	0.045	15	1.0067	15	
	LFS 2 0602 (0-10') rem. on 1/4" & #40	0.051	74	1.0058	74	
	LFS 2 0602 (10-20') all bio. fine ground	0.048	90	1.0076	89	
	LFS 2 0604 (15-30') pass #40	0.036	24	1.0055	24	
	CCB	0.018	1			
	CCV (50 ng Hg)	0.040	40			

Table A.1 (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb. (253.5 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)
3/17/96	LFS 2.0804 (10-20') rem. org. on 1/4" & #40	0.060	104	1.0071	105	
	LFS 2.0804 (20-30') pass. #40	0.105	107	1.0058	106	
	LFS 2.0602 (20-30') rem. on 1/4" & #40	0.254	548	1.0070	544	
	LFS 2.0602 (0-10') rem. org. on 1/4" & #40	0.063	79	1.0040	78	
	LFS 2.0901 (0-10') rem. org. on 1/4" screen	0.025	17	1.0027	17	
	LFS 2.0902 (10-20') rem. org. on 1/4" screen	0.177	361	1.0063	358	
	LFS 2.0902 (30-40') rem. on 1/4" screen	0.161	370	1.0068	367	
	LFS 2.0902 (40-45') rem. org. on 1/4" screen	0.066	99	1.0015	99	
	LFS 2.0903 (35-45') rem. on 1/4" screen	0.118	222	1.0068	221	
	LFS 3.0901 (10-20') rem. on 1/4" screen	0.450	1051	1.0020	1049	Redo
	LFS 3.0901 (20-30') 1/4" screen removed	0.081	142	1.0077	141	
	LFS 3.0901 (30-40') rem. on 1/4" screen	0.036	49	1.0034	49	
	LFS 3.0901 (40-45') rem. on 1/4" screen	0.400	868	1.0038	865	Redo
	CCB	0.016	0	1.0116	0	
	CCV (50 ng Hg)	0.036	47	1.0067	46	
	LFS 3.0902 (10-20') rem. on 1/4" screen			1.0036		Redo
	LFS 3.0902 (20-30') rem. on 1/4" screen			1.0077		Redo
	LFS 3.0902 (30-40') rem. org. on 1/4" screen			1.0082		Redo
	LFS 3.0902 (40-45') rem. on 1/4" screen	0.112	213	1.0061	212	
	LFS 3.0902 (40-45') rem. on 1/4" screen (DUP)	0.115	220	1.0046	218	
	LFS 4.0801 (10-20') pass #40	0.039	47	1.0031	47	
	LFS 4.0801 (10-20') rem. on 1/4" & #40	0.066	104	1.0017	103	
	LFS 4.0801 (20-25') pass #40	0.049	60	1.0031	60	
	LFS 4.0801 (20-25') rem. org. on 1/4" & #40	0.144	286	1.0074	284	
	LFS 4.0802 (10-20') all blo. ground fine	0.031	20	1.0007	20	
	CCB	0.018	0			
	CCV (50 ng Hg)	0.039	47			
	LFS 4.0802 (10-20') pass #40	0.037	42	1.0006	42	
	LFS 4.0802 (10-20') rem. on 1/4" & #40	0.040	49	1.0042	49	

Table A.1 (con't d) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% recy)
	LFS 4 0802 (20-30') all b/o ground fine	0.117	224	1.0006	224	
	LFS 4 0802 (20-30') pass #40	0.068	113	1.0045	112	
	LFS 4 0802 (20-30') pass #40 (SP)	0.060	154	1.0042	153	81.9
	LFS 4 0802 (20-30') rem. org on 1/4" & #40	0.113	216	1.0085	214	
	CCB	0.018	3			
	CCV (50 ng Hg)	0.040	48			

Table A-1 (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D	Absorb (253.7 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
4/26/96	Blank	0.024	0.00				
	Std 1 (50 ng)	0.043	60.00				
	Std 2 (150 ng)	0.085	147.77				
	Std 3 (250 ng)	0.131	264.00				
	Std 4 (350 ng)	0.194					
	Std 5 (450 ng)	0.198	423.10				
	Std 6 (500 ng)	0.251	567.22				
	Check Std	0.079	130.00	1.0160	128		
	LFS 5 #401 (0-10')	0.031	11.32	1.0014	11	32/96	34
	LFS 5 #401 (0-10') pass #40	0.048	97.23	1.0016	97		
	LFS 5 #401 (0-10') pass #40 (DUP)	0.367	102.00	1.0012	102		
	LFS 5 #401 (0-10') rem. on #40	0.048	71.00	1.0016	72	33/33	144
	LFS 5 #401 (10-20')	0.072	114.92	1.0040	114		
	LFS 5 #401 (10-20') pass. #40	0.061	61.65	1.0025	62		
	LFS 5 #401 (10-20') rem on #40	0.061	61.65	1.0014	59	33/33	124
	LFS 5 #401 (20-30') pass. #40	0.097	178.00	1.0058	177		
	LFS 5 #401 (20-30') pass #40 (SP)	0.115	223.57	1.0074	222	91.0	
	LFS 5 #401 (20-30') rem on #40	0.220	509.11	1.0016	508	33/28	1010
	LFS 5 #401 (30-40') pass #40	0.066	104.01	1.0027	104		
	LFS 5 #401 (20-30') rem. on #40	0.111	213.48	1.0058	212	39/36	424
	CCB	0.024	0.00				
	CCV (50 ng Hg)	0.049	60.00				
	LFS 5 #402 (0-10')	0.199	425.72	1.0038	424	33/66	1272
	LFS 5 #402 (0-10') pass. #40	0.440	1044.00	1.0043	1040	Redo	
	LFS 5 #402 (10-20') pass. #40	0.075	122.90	1.0010	122		
	LFS 5 #402 (10-20') rem on #40	0.072	114.92	1.0024	115	33/39	229
	LFS 5 #402 (20-30') pass #40	0.247	557.12	1.0001	555		
	LFS 5 #402 (20-30') rem on #40	0.142	691.00	1.0065	640	30/36	1100
	LFS 5 #402 (0-10') org. mat	0.040	34.00	1.0122	34		

Table A.1 (cont'd) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb. (253.5 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
	LFS 5.0403 (0-10') pass. #40	0.048	41.84	1.0061	41		
	LFS 5.0403 (10-20') pass. #40			1.0080		Redo	
	LFS 5.0403 (10-20') pass. #40 (DUP)			1.0487		Redo	
	LFS 5.0403 (0-10') rem. on #40	0.027	1.21	1.0013	1	33/33	2
	LFS 5.0403 (10-20') org. & rest	0.062	55.96	1.0054	56	20/80	367
	CCB	0.043	0.00				
	CCV (50 ng Hg)	0.049	50.60				
	LFS 5.0403 (10-20') rem. on #40			1.0019		Redo	
	LFS 5.0403 (20-30') org. & rest	0.066	94.70	1.0451	94		
	LFS 5.0403 (20-30') pass. #40	0.091	160.93	1.0009	161		
	LFS 5.0403 (20-30') rem. on #40	0.056	71.90	1.0018	72	20/80	287
	LFS 5.0403 (30-40')	0.056	84.38	1.0005	84		
	LFS 5.0403 (30-40') pass. #40	0.067	100.28	1.0048	100		
	LFS 5.0403 (30-40') rem. on #40	0.065	49.22	1.0017	49	33/33	98
	LFS 5.0701 (5-7.5') pass. #40	0.055	18.37	1.0027	18		
	LFS 5.0701 (5-7.5') rem. on 1/4" & #40	0.060	74.43	1.0000	74	20/80	288
	LFS 5.0701 (15-17.5') pass. #40			1.0012		Redo	
	LFS 5.0701 (35-37.5') rem. on 1/4" & #40	0.099	175.00	1.0013	175		
	LFS 5.0701 (25-27.5') rem. on 1/4" & #40	0.081	81.06	1.0020	81	20/80	247
	CCB	0.024	0.00				
	CCV (60 ng Hg)	0.062	59.33				
	LFS 5.0702 (0-10') pass. #40	0.041	36.56	1.0011	37		
	LFS 5.0703 (0-10') pass #40	0.041	36.56	1.0015	37		
	LFS 5.0703 (0-10') rem. on 1/4" & #40	0.072	114.92	1.0010	115		
	LFS 5.0703 (30-40') pass. #40	0.183	310.56	1.0024	310		
	LFS 5.0702 (30-40') pass. #40 (DUP)	0.188	327.17	1.0001	327		
	CCB	0.025	0.00				
	CCV (60 ng Hg)	0.048	54.27				

Table A-1 (con't) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb. (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
5/6/98	Blank	0.023	2				
	Std 1 (50 ng)	0.044	55				
	Std 2 (150 ng)	0.079	151				
	Std 3 (250 ng)	0.110	238				
	Std 4 (350 ng)	0.148	341				
	Std 5 (450 ng)	0.186	445				
	Std 6 (550 ng)	0.230	564				
	Check Std	0.070	129	1.0022	129		
	LFS 3 0901 (10-20') bio. rem. on #40	0.151	216	1.0084	213	33/33	626
	LFS 3 0901 (20-30') bio. rem. on #40	0.159	371	1.0039	370	22/44	1106
	LFS 3 0901 (30-40') bio. rem. on #40	0.068	96	1.0065	96	22/44	285
	LFS 3 0901 (40-45') bio. rem. on #40	0.170	401	1.0068	399	25/25	787
	LFS 3 0902 (10-20') bio. rem. on #40	0.144	331	1.0020	330	25/25	660
	LFS 3 0902 (20-30') bio. rem. on #40	0.134	309	1.0013	308	25/25	608
	LFS 3 0902 (30-40') bio. rem. on #40	0.143	329	1.0084	326	5/45	3264
	LFS 3 0902 (40-45') bio. rem. on #40	0.080	157	1.0032	156	5/45	1561
	LFS 5 5701 (35-37.5') pass. #40	0.047	67	1.0045	67	25/25	153
	LFS 5 5701 (25-27.5') pass. #40	0.077	149	1.0038	148	25/25	595
	LFS 5 5705 (35-38') pass. #40	0.040	44	1.0070	44		
	LFS 5 5705 (35-38') pass. #40 (DUP)	0.044	56	1.0060	56		
	LFS 5 0705 (35-38') pass. #40 (SP)	0.080	106	1.0078	101	98.9	
	CCB	0.023	2				
	CCV (50 ng Hg)	0.042	53				
	LFS 5 0901 (5-15') bio. rem. on #40	0.067	94	1.0024	94	33/33	186
	LFS 5 0901 (15-25') rem. on #40	0.064	113	1.0053	113	25/25	320
	LFS 5 0901 (25-35')A bio. rem. on #40	0.142	325	1.0062	323	33/33	840
	LFS 5 0901 (35-45') bio. rem. on #40	0.203	401	1.0047	488	33/33	977
	LFS 5 0901 (25-35')B bio. rem. on #40	0.120	266	1.0027	265	33/33	588
	LFS 5 0902 (5-15') bio. rem. on #40	0.068	124	1.0062	123	33/33	247

Table A-1 (con't d) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb. (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
	LFS 6 0502 (15-25') blc: mm on #40	0.048	70	1.0061	69	33/33	139
	LFS 5 0502 (25-35') blc: mm on #40	0.176	417	1.0026	416	11/55	2496
	LFS 5 0502 (35-45') blc mm on #40	0.042	53	1.0065	53	11/55	318
	CCB	0.023	2				
	CCV (50 ng Hg)	0.043	56				
	Redos Below						
	LFS 1 0502 (0-15')A3rd mm on #40	0.180	428	1.0006	427	25/25	854
	LFS 1 0901 (25-35') blc mm on #40	0.414	1064	1.0047	1059	0.5/55.5	99333
	LFS 1 0901 (35-45') mm on #40	0.137	311	1.0098	310	20/80	1238
	LFS 1 0902 (25-35') blc: mm on #40	0.065	88	1.0024	88	11/55	531
	LFS 2 0301 (10-20')D pass #40	0.154	358	1.0028	357	25/25	713
	LFS 2 0801 (20-30') all blc. fine ground	0.044	70	1.0067	69	11/55	416
	LFS 2 0902 (30-40') mm on #40	0.181	431	1.0021	430	25/25	860
	LFS 3 0901 (20-30') 1/4" bar removed	0.080	187	1.0053	186	25/50	323
	LFS 3 0001 (40-45') mm on 1/4"	0.117	257	1.0067	256	20/80	1023
	LFS 3 0902 (10-20') mm 1/4"	0.081	109	1.0092	108	45/45	209
	LFS 3 0902 (20-30') mm 1/4"	0.200	480	1.0058	478	20/80	1910
	LFS 3 0502 (20-30') mm 1/4"	0.116	254	1.0070	252	20/80	1010
	LFS 5 0402 (0-10') pass #40	0.200	483	1.0098	481	25/25	960
	LFS 6 0403 (10-12') pass. #40	0.083	165	1.0038	164	25/25	328
	LFS 5 0403 (10-20') mm on #40	0.077	148	1.0074	147	11/55	690
	LFS 5 0502 (25-35') mm on 1/4"	0.147	339	1.0063	337	45/45	674
	CCB	0.023	2				
	CCV (50 ng Hg)	0.042	53				

Table A.1 (cont. 5) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
5/3/06	Blank	0.018	0				
	Std 1 (50 ng)	0.037	56				
	Std 2 (150 ng)	0.073	148				
	Std 3 (250 ng)	0.115	237				
	Std 4 (350 ng)	0.152	342				
	Std 5 (450 ng)	0.198	461				
	Std 6 (550 ng)	0.240	561				
	Check Std	0.075	138	1.0190	136		
	LFS 5 0704 (0-10') pass #40	0.182	441	1.0079	438		
	LFS 5 0704 (0-10') pass #40 (DUP)	0.180	411	1.0074	408		
	LFS 5 0704 (0-10') rem. on 1/4" & #40	0.108	232	1.0044	231	20/50	778
	LFS 5 0704 (10-20') pass #40	0.115	260	1.0070	258		
	LFS 5 0704 (10-20') rem. on 1/4" & #40	0.084	185	1.0053	184	20/50	657
	LFS 5 0704 (20-30') pass #40	0.056	108	1.0030	107		
	LFS 5 0704 (20-30') pass #40 (SP)	0.080	185	1.0034	182	139.5	
	LFS 5 0704 (20-30') rem. 1/4" & #40	0.031	41	1.0037	40	20/60	103
	LFS 5 0704 (30-40')A pass #40	0.203	408	1.0074	405		
	LFS 5 0704 (30-40')A rem. 1/4" & #40	0.06	116	1.0088	115	20/60	460
	LFS 5 0704 (30-40')B pass #40	0.150	337	1.0066	335		
	LFS 5 0704 (30-40')B rem. 1/4" & #40	0.127	280	1.0027	279	30/33	568
	LFS 5 0705 (0-7.5') pass #40	0.041	65	1.0048	65		
	LFS 5 0704 (5-7.5') rem. 1/4" & #40	0.033	46	1.0071	46	20/60	161
	CCB	0.010	0				
	CCV (50 ng Hg)	0.050	53				
	LFS 5 0705 (15-17.5') pass #40	0.055	100	1.0050	100		
	LFS 5 0705 (15-17.5') rem. 1/4" & #40	0.057	105	1.0080	104	20/50	410
	LFS 5 0704 (25-27.5') pass #40	0.152	342	1.0045	340		
	LFS 5 0705 (25-27.5') rem. 1/4" & #40	0.050	61	1.0080	60	20/50	241
	LFS 5 0705 (35-36') rem. 1/4" & #40	0.096	202	1.0020	202	33/33	404

Table A-1 (con't d) Raw Mercury Data for Alachua County Landfill Residues

Date	Sample I.D.	Absorb (253.6 nm)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)	Comments (% rec'y)	[Hg] (ng/g)
	LFS 5 0901 (5-15') nm 1/4" screen	0.038	61	1.0041	61		
	LFS 5 0901 (15-25') nm. 1/4" screen	0.094	197	1.0096	195		
	LFS 5 0901 (25-35') nm 1/4" screen	0.155	349	1.0043	348		
	LFS 5 0901 (35-45') nm. 1/4 screen	0.135	209	1.0010	209		
	LFS 5 0902 (5-15') nm 1/4" screen	0.054	98	1.0040	97		
	LFS 5 0902 (15-25') nm 1/4 screen	0.048	75	1.0028	75		
	LFS 5 0902 (25-35') nm 1/4 screen	0.268	633	1.0051	630	Redo	
	LFS 5 0902 (25-35') nm. 1/4 screen			1.0056		Redo	
	LFS 5 0902 (35-45') nm. 1/4 screen	0.048	63	1.0051	63	25/50	251
	CCB	0.018	8				
	CCV (50 ng Hg)	0.034	48				
5/8/96	LFS 1 0901 (5-15') b/e. nm on #40	0.118	252	1.0018	252		
	LFS 1 0901 (15-25') b/e nm. on #40	0.238	252	1.0016	252		
	LFS 1 0901 (25-35') b/e. nm. on #40		551	1.0087	547	Redo	
	LFS 1 0901 (35-45') b/e nm on #40	0.026	33	1.0040	33	1/50	1650
	LFS 1 0902 (5-15') b/e nm on #40	0.136	295	1.0010	295		
	LFS 1 0902 (15-25') b/e nm. on #40	0.106	227	1.0007	227		
	LFS 1 0902 (25-35') b/e. nm. on #40			1.0033		Redo	
	LFS 1 0902 (35-45') b/e nm on #40	0.074	148	1.0057	147	25/25	448
	LFS 1 0902 (5-15') b/e nm on #40	0.069	125	1.0063	125	25/25	248
	LFS 1 0903 (15-25') b/e nm on #40	0.087	180	1.0032	179	33/33	358
	LFS 1 0902 (25-35') b/e. nm. on #40	0.131	290	1.0024	289	33/33	578
	LFS 1 0902 (35-45') b/e nm. on #40	0.077	155	1.0069	154	25/25	308
	CCB	0.017	0				
	CCV (50 ng Hg)	0.036	53				
	LFS 2 0901 (0-10') b/e nm on #40	0.067	130	1.0040	130	33/33	259
	LFS 2 0901 (10-20') b/e. nm. on #40	0.188	282	1.0062	280	33/30	561
	LFS 2 0901 (20-30') b/e nm. on #40	0.188	424	1.0034	422	10/40	2112
	LFS 2 0901 (20-30') b/e nm. on #40 (DUP)	0.190	411	1.0481	410	10/40	2049

APPENDIX B
RAW DATA FOR ANAEROBIC REACTORS

Table B-1 pH Raw Data for Run 1

Day	R1	R2	R3	R4	R5	R6	R7	R8	R9
0	7.99	7.59	7.98	7.59	7.50	7.88	7.55	7.88	7.88
1	7.78	7.83	7.88	7.86	7.80	7.76	7.78	7.83	7.53
2	7.38	7.41	7.20	7.36	7.89	7.53	7.14	7.32	7.41
3	6.99	7.08	7.02	7.02	7.11	6.98	6.90	6.87	6.84
4	6.84	5.8	6.53	6.95	7.34	6.80	6.94	7.04	7.08
5	6.78	6.71	6.76	6.67	6.81	6.83	6.71	6.78	6.58
7	7.12	7.13	7	7.01	7.34	7.12	7	7.08	7.07
9	7.61	7.64	7.90	7.68	7.45	7.54	7.35	7.56	7.61
11	7.6	7.93	7.60	7.84	7.71	7.64	7.80	7.88	7.93
13	7.64	7.87	7.50	7.75	7.75	7.77	7.88	7.77	7.7
15	7.64	8.03	7.99	8.09	8.00	7.67	7.53	7.76	7.53
17	7.7	7.94	7.86	8.03	7.98	7.90	7.9	7.83	7.83
19	7.98	7.8	8.03	8.08	7.99	8.05	7.88	7.93	7.88
22	7.93	7.8	7.84	8.04	7.95	8.05	7.93	7.96	7.98
29	7.74	7.8	8.07	8.01	7.94	8.01	7.88	7.99	7.09
31	7.74	7.09	8	8.11	8.13	8.07	7.91	8.20	8.15

Table II-2. pH Raw Data for Run 2

Day	R1	R2	R3	R4	R5	R6	R7	R8	R9
0	7.61	7.61	7.61	7.61	7.61	7.61	7.61	7.61	7.61
1	7.58	7.3	7.55	7.61	7.35	7.45	7.4	7.46	7.54
2	7.34	7.08	7.64	7.16	7.2	7.56	7.22	7.22	7.43
3	6.96	6.61	6.60	6.78	7	6.74	6.89	7.56	7.16
4	7.56	7.02	7.08	7.31	7.45	7.4	7.25	7.56	7.18
5	7.38	7.30	7.43	7.34	7.56	7.38	7.2	7.63	7.09
6	7.66	7.65	7.76	7.76	7.56	7.67	7.76	8.03	7.55
7	7.83	7.78	8.05	8	7.96	8	8.1	8.16	7.94
8	8.18	7.89	7.94	7.88	8.16	7.92	8.06	7.81	7.94
10	7.56	7.93	7.66	7.47	7.88	7.45	7.36	8.01	7.39
12	8.16	7.96	7.87	8.18	8.16	8.18	8.12	8.07	8.18
14	8.01	7.6	8.03	8.06	8.06	8.01	8.03		8.06

Table B-3 Methane % Data for Run 1

Day	R1	R2	R3	R4	R5	R6	R7	R8	R9
0	0.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.04
1	2.00	0.28	0.00	0.06	0.00	0.00	0.00	0.06	0.02
2	5.66	0.32	0.00	0.78	0.56	1.18	1.00	1.02	2.67
3	17.58	11.29	6.33	11.33	3.78	6.07	8.54	12.00	17.50
4	25.62	22.17	16.92	17.97	21.80	17.16	24.32	21.80	55.58
5	39.17	23.26	27.22	30.34	33.07	23.96	31.78	34.36	42.84
7	56.06	47.36	42.73	41.90	25.42	43.15	43.36	44.16	56.18
8	53.67	53.64	49.91	52.05	48.96	44.75	54.13	53.00	54.00
9	56.31	58.70	48.62	48.02	54.90	46.87	58.77	60.58	56.43
10	56.18	57.13	52.12	52.39	59.85	60.85	46.74	54.41	56.18
11	54.90	55.51	56.34	53.41	56.69	49.16	47.56	51.00	56.58
12	54.61	56.40	52.44	47.90	51.09	46.11	52.23	60.76	52.00
13	54.31	54.06	50.60	53.01	62.34	48.54	40.36	55.09	52.80
15	57.70	58.01	51.35	53.28	55.22	54.96	47.39	58.73	59.43
16	54.34	57.80	52.86	60.65	56.30	47.67	48.54	51.78	56.84
17	56.63	58.27	54.16	43.53	54.28	61.43	56.11	48.72	55.21
18	59.40	60.46	53.76	50.97	62.46	43.98	46.00	50.05	56.38
19	57.96	60.83	53.13	48.08	60.69	53.16	43.37	51.09	42.97
22	56.01	60.70	53.02	49.48	37.56	56.09	44.80	42.06	30.27
25	56.26	58.84	56.12	48.09	54.71	53.28	47.08	56.00	60.81
26	57.56	52.33	60.46	60.60	54.09	42.81	47.38	58.59	58.03
28	56.58	60.76	57.28	50.50	54.88	45.60	45.20	48.75	61.84
31	59.06	58.84	58.49	48.91	57.13	50.44	60.27	60.27	58.78
35	59.06	44.00	57.36	41.84	52.50	54.61	51.93	58.13	58.10

Table 5-4 Raw Methane % Data for Run 7

Date	Day	R1	R2	R3	R4	R5	R6	R7	R8	R9
7 May	1	37 77	71 77	33 77	77 77	79 77	14 94	72 84	33 77	77 77
7 May	7	77 77	47 77	75 77	77 77	47 77	77 12	78 97	47 77	77 77
7 May	3	77 77	77 78	77 77	77 77	77 77	77 77	42 77	77 77	81 77
7 May	7	77 78	77 77	77 36	77 17	45 77	77 77	77 77	77 77	78 04
8 May	5	77 17	77 77	77 22	77 77	77 71	77 77	77 73	77 77	52 56
7 May	8	77 77	77 77	51 29	72 71	77 77	77 72	44 47	77 81	77 07
7 May	7	77 77	77 77	77 22	51 71	77 77	77 72	77 77	77 72	77 47
7 May	8	71 78	42 78	77 77	78 51	77 77	77 77	77 77	78 71	53 47
71 May	77	77 70	77 77	54 77	50 06	59 77	77 77	77 77	77 82	77 77
13 May	77	54 78	77 27	54 54	56 87	77 77	78 77	77 77	77 57	47 77
77 May	77	54 10	77 78	77 80	78 78	77 77	77 77	77 77	77 80	77 77
77 May	17	54 78	77 78	77 73	55 64	77 77	65 77	77 77	77 77	77 77
19 May	17	54 67	75 19	54 72	88 76	55 77	54 77	75 78	77 27	77 37

Table B-5. Methane Yield (m^3/kg VS added) for Run 1

Day	R1	R2	R3	R4	R5	R6	R7	R8	R9
0	0.0001035	0	0	0	0	0	0	0	0
1	0.0005513	4.602E-05	6.798E-06	0	0	0	0	0	[illegible]
2	0.0010681	7.175E-06	1.849E-07	0.0001397	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
3	0.0073925	0.0021097	0.0011943	0.0020317	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
4	0.0047619	0.0040991	0.0034628	0.0032541	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
5	0.0072424	0.0051717	0.0050839	0.0055532	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
7	0.0102569	0.0087130	0.0080223	0.0077524	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
8	0.0058565	0.0086653	0.0090195	0.0096673	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
9	0.0080244	0.0094285	0.0074205	0.0082318	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
10	0.0069052	0.0089486	0.0079659	0.0087073	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
11	0.0066122	0.0071217	0.0064309	0.0059525	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
12	0.0067217	0.0073426	0.0061165	0.0053283	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
13	0.0056364	0.0060166	0.0051635	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
15	0.0050078	0.0045859	0.0042144	0.0040714	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
16	0.0047166	0.0044016	0.0043487	0.0031443	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
17	0.0030673	0.0036884	0.0034750	0.0026204	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
18	0.0041605	0.0034747	0.0035151	0.0031256	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
19	0.0038249	0.0035522	0.0035006	0.0024950	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
22	0.0090889	0.0097551	0.0100003	0.0077235	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
25	[illegible]	0.0038513	0.0042358	0.0031676	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
26	0.0035837	0.0037628	0.0041871	0.0037355	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
29	0.0054357	0.0038556	0.0103749	0.0091658	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
31	0.0074282	0.0040165	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
36	0.0050718	0.0040332	0.0045785	0.0035521	[illegible]	0.0051066	[illegible]	[illegible]	0.00117

Table B-6 Methane Yield (m^3/kg VS added) for Run 1

Days	R1	R2	R3	R4	R5	R6	R7	R8	R9
1	1 1110704	1 1111111	1 1164111	0 1153110	0 0041111	1 1182111	1 1118361	1 1158110	1 1111166
1	0 1011172	1 1111821	1 1111117	0 0121146	1 0091711	1 1111671	1 1131111	1 1131111	1 1161111
1	0 0137011	1 1111111	1 1111111	1 9811982	1 1113131	1 8282111	1 1131311	1 1111131	0 1051011
4	0 0171111	1 1111111	1 1111111	1 1111949	1 1116120	1 1212711	1 0111212	1 1111111	1 0111261
5	1 1111111	1 1117515	1 1111111	1 1111111	1 1116111	1 8211111	0 0221118	1 1211131	1 1141111
1	1 1103611	1 1111110	1 1111111	1 1111111	1 8211824	0 1118317	0 0175188	0 1111541	1 1111101
7	1 1132111	1 1111141	1 1111111	1 1111111	1 1211291	1 1111981	0 0111154	0 1111111	1 1111111
1	1 1141122	1 1110817	1 1111111	1 1111112	1 1111111	1 0160485	0 0111741	1 0111111	1 1111113
11	1 1111111	1 0201101	1 8211231	1 1115111	1 1111111	1 1116604	1 8211111	1 8211111	1 1111116
13	1 8211111	0 1277281	0 8211111	1 1111111	1 1111111	1 1111111	1 1111111	1 1161011	1 1111821
11	1 1177131	1 0180158	0 0111	1 1111111	1 1111111	1 1111111	1 1111111	1 1111111	1 1111751
11	1 1115541	0 0126111	1 0135353	1 1111111	1 0111182	1 1111111	1 1111111	1 1111156	0 1081301
10	1 1821117	0 111111	1 1191919	1 1111110	0 0091111	1 1111111	1 1111110	1 1114117	1 9082111

APPENDIX C
RAW MERCURY DATA FOR ANAEROBIC REACTORS

Table 6-1 Run 1 Reactor 1 Solids Data

Sample I D	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
Blank	0.017	0		
Std 1 (20 ng)	0.027	20		
Std 2 (40 ng)	0.036	38		
Std 3 (80 ng)	0.055	76		
Std 4 (100 ng)	0.065	96		
Std 5 (200 ng)	0.110	186		
Std 6 (300 ng)	0.160	286		
IDB	0.016	0		
MH	0.017	0		
SP(40 ng)	0.036	38		
R1:1	0.017	0	2.0997	0
R1:2	0.019	4	2.1330	10
R1:3	0.016	0	2.2512	0
R1:4	0.015	0	2.2474	0
R1:5	0.017	0	2.1106	0
R1:6	0.017	0	2.1049	0
R1:7	0.017	0	2.0774	0
R1:8	0.017	0	2.2601	0
R1:9	0.017	0	2.0008	0
R1:10	0.016	0	2.1192	0
R1:11	0.019	4	2.3171	9
R1:12	0.017	0	2.0248	0
R1:13	0.017	0	2.3400	0
R1:14	0.017	0	2.0580	0
R1:15	0.017	0	2.0072	0
R1:16	0.015	0	2.1603	0
R1:17	0.015	0	2.0060	0
R1:18	0.017	0	2.2206	0
R1:19	0.015	0	2.0230	0
R1:20	0.017	0	2.0642	0
R1:21	0.016	0	2.1972	0
R1:22	0.017	0	2.2033	0
R1:23	0.015	0	2.1907	0
R1:24	0.016	0	2.1687	0
R1:25	0.017	0	2.3480	0
R1:26	0.015	0	2.1283	0
R1:27	0.015	0	2.1135	0
R1:28	0.014	0	2.2808	0
R1:29	0.017	0	2.1548	0
R1:30	0.016	0	2.1546	0
R1:31	0.014	0	2.1522	0
R1:32	0.017	0	2.1597	0
R1:33	0.015	0	2.1026	0
R1:34	0.017	0	2.0898	0

Constant:	0.0171
X Coef..	0.0005
Solids (g/g)	0.1861

Table C-1 Run 1 Reactor 1 Solids Data

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt grams	[Hg] (ng/g)
R1:35	0.017	0	2.0508	0
R1:36	0.018	0	2.2468	0
R1:37	0.018	0	2.1327	0
R1:38	0.018	0	2.2600	0
R1:39	0.017	0	2.1495	0
R1:40	0.018	0	2.1586	0
R1:41	0.017	0	2.2640	0
R1:42	0.017	0	2.0508	0
R1:43	lost in bath			
R1:44	0.018	0	2.2929	0
R1:45	0.018	0	2.3062	0
R1:46	0.017	0	2.1860	0
	Sum	0	Sum	16

Table C-2. Run 1 Reactor 4 (Solids)

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
Blank	0.000	0		
Std 1 (50 ng)	0.006	40		
Std 2 (100 ng)	0.066	92		
Std 3 (200 ng)	0.080	175		
Std 4 (300 ng)	0.135	306		
Std 5 (400 ng)	0.170	396		
Std 6 (500 ng)	0.210	475		
Std 7 (600ng)	0.255	585		
Std 8 (800 ng)	0.333	753		
ICB	0.021	0		
MB	0.021	0		
SP(50 ng)	0.038	45		
R4 1	0.021	0	2.2520	0
R4:2	0.020	0	2.3044	0
R4:3	0.020	0	2.1067	1
R4:4	0.020	0	2.1268	1
R4:5	0.018	0	2.2484	0
R4:6	0.021	0	2.1213	0
R4:7	0.019	0	2.2756	0
R4:8	0.021	0	2.3444	0
R4:9	0.008	0	2.2258	0
R4:10	0.015	0	2.3364	0
R4:11	0.018	0	2.1866	0
R4 12	0.023	0	2.2734	16
R4 13	0.006	0	2.0508	0
R4 14	0.018	0	2.3432	0
R4 15	0.021	0	2.1235	0
R4 16	0.020	0	2.1227	1
R4 17	0.020	0	2.2089	0
R4 18	0.017	0	2.1303	0
R4 19	0.022	0	2.0565	0
R4 20	0.020	0	2.2742	0
R4:21	0.021	0	2.4045	0
R4:22	0.022	0	2.0026	0
R4 23	0.006	0	2.3605	0
R4:24	0.021	0	2.0927	0
R4 25	0.020	0	2.0951	0
R4:26	0.008	0	2.1487	1
R4:27	0.023	0	2.3488	14
R4:28	0.020	0	2.1857	0
R4 29	0.021	0	2.1248	0
R4 30	0.021	0	2.0814	0
R4 31	0.018	0	2.3298	0
R4:32	0.020	0	2.3566	0

Constant:	0.0147
X Coef	0.0004
Solids (g/g)	0.2512

Table C-2 (cont'd) Run 1 Reactor 4 (Solids)

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
R4.33	0.023	8	2.1280	18
R4.34	0.018	0	2.2342	0
R4.35	0.020	0	2.3686	0
R4.36	0.021	0	2.1612	0
R4.37	0.014	0	2.2184	0
R4.38	0.017	0	2.3134	0
R4.39	0.022	0	2.2358	0
R4.40	0.031	8	2.2983	17
R4.41	0.018	0	2.2601	0
R4.42	0.017	0	2.1621	0
R4.43	0.021	0	2.2098	0
R4.44	0.032	0	2.3628	0
R4.45	0.030	0	2.0839	1
R4.46	0.021	0	2.3664	0
R4.47	0.028	20	2.3681	37
R4.48	0.017	0	2.1918	0
R4.49	0.021	0	2.4891	0
R4.50	0.021	0	1.8379	0
R4.51	0.020	7	1.5218	1
	S =	56	S =	100

Table C-3. Run 1 Reactor 7 (Solids)

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
Blank	0.021	0		
Std 1 (50 ng)	0.043	44		
Std 2 (100 ng)	0.067	92		
Std 3 (200 ng)	0.123	202		
Std 4 (400 ng)	0.210	378		
Std 5 (600 ng)	0.324	606		
Std 6 (800 ng)	0.410	770		
Std 7 (1000ng)	0.470	896		
ICB	0.021	0		
MS	0.021	0		
SP(50 ng)	0.048	46		
R7:1	0.033	24	2.1642	52
R7:2	0.041	38	2.1145	86
R7:3	0.028	14	2.1744	30
R7:4	0.021	0	2.0930	1
R7:5	0.027	12	2.1606	27
R7:6	0.021	0	2.1628	1
R7:7	0.021	0	2.1583	1
R7:8	0.015	0	2.3585	0
R7:9	0.031	20	2.1364	45
R7:10	0.021	0	2.1357	1
R7:11	0.023	0	2.5004	0
R7:12	0.021	0	2.0631	1
R7:13	0.055	68	2.2419	146
R7:14	0.023	0	2.4749	0
R7:15	0.225	406	2.3166	841
R7:16	0.020	0	2.0110	0
R7:17	0.164	286	2.0861	674
R7:18	0.020	0	2.0217	0
R7:19	0.020	0	2.1666	0
R7:20	0.021	0	2.3208	1
R7:21	0.020	0	2.1908	0
R7:22	0.021	0	2.1354	1
R7:23	0.036	18	2.2057	62
R7:24	0.142	242	2.3713	490
R7:25	0.023	0	2.0361	0
R7:26	0.026	10	2.1406	20
R7:27	0.033	24	2.3526	49
R7:28	0.024	0	2.0944	0
R7:29	0.048	56	2.2484	122
R7:30	0.036	28	2.0530	65
R7:31	0.072	99	2.3653	202
R7:32	0.026	9	2.0683	20
R7:33	0.046	50	2.2645	84

Constant	0.0245
X Coef	0.0005
Solids (g/g):	0.2084

Table C-3 (cont'd). Run 1 Reactor 7 (Solids)

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
R7:34	0.054	26	2.4128	53
R7:35	0.024	0	2.4265	0
R7:36	0.023	0	2.3683	0
R7:37	0.020	0	2.1060	0
R7:38	0.022	0	2.2330	0
R7:39	0.146	325	2.2544	715
R7:40	0.026	14	2.3256	30
R7:41	0.021	0	2.4785	1
R7:42	0.019	0	2.2832	0
R7:43	0.020	0	1.5144	0
R7:44	0.020	0	1.3856	0
	Σ =	1767	Σ =	3860

Table C-4 Run 1 Leachate Raw Data

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt ml	[Hg] (ng/g)
Blank	0.033	0		
Std 1 (10 ng)	0.037	9		
Std 2 (30 ng)	0.043	25		
Std 3 (50 ng)	0.054	52		
Std 4 (80 ng)	0.065	80		
Std 5 (100 ng)	0.073	100		
Std 6 (200 ng)	0.118	206		
Std 7 (300ng)	0.150	296		
R1	0.033	0	100	
R2	0.033	0	100	
R3	0.025	0	98	
R4	0.038	12	100	
R5	0.024	0	100	
R6	0.019	0	100	
R7	0.023	0	100	
R8	0.023	0	100	
R8 (dup)	0.022	0	100	
R9	0.023	0	100	
	S =	12		

Constant:	0.0332
X Coef.	0.0004
Solids (g/g):	0.0094

Table C-5 Run 1 Activated Carbon Data

Constant: 0.0162
X Coef: 0.0006

Sample I.D.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
Blank	0.018	0		
Std 1 (5 ng)	0.021	8		
Std 1 (10 ng)	0.020	10		
Std 2 (20 ng)	0.030	24		
Std 3 (30 ng)	0.033	30		
Std 4 (50 ng)	0.044	52		
Std 5 (60 ng)	0.053	70		
Std 6 (100 ng)	0.060	102		
Std 7 (200 ng)	0.102	162		
Std 8 (300ng)	0.184	292		
Std 9 (500 ng)	0.257	476		
ACs 1	0.016	0		
ACs 2	0.018	0		
ACs 3	0.017	0		
ACs 4	0.027	18		
ACs 5	0.020	6		
ACs 6	0.027	18		
ACs 7	0.030	24		
ACs 8	0.033	30		
ACs 9	0.036	34		
	S =	102		

Table C-6. Run 1 Tygon Tubing Data

Sample I D	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
Blank	0.002	0		
Std 1 (10 ng)	0.026	8		
Std 3 (30 ng)	0.033	22		
Std 4 (50 ng)	0.044	44		
Std 5 (80 ng)	0.054	64		
Std 6 (100 ng)	0.067	90		
Std 7 (200 ng)	0.112	180		
R1	0.030	0		
R2	0.023	0		
R3	0.022	0		
R4	0.038	12		
R5	0.058	28		
R6	0.030	10		
R7	0.035	26		
R8	0.031	18		
R9	0.028	8		
	Σ =	106		

Constant:	0.0214
X Coef.	0.0006

Table C-7 Run 2 Reactor 1 Solids Data

Sample I.D.	Absorbance (253 ß)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
Blank	0.034	0		
Std 1 (10 ng)	0.009	6		
Std 2 (30 ng)	0.040	30		
Std 3 (50ng)	0.067	46		
Std 4 (80 ng)	0.067	56		
Std 5 (100 ng)	0.076	64		
Std 6 (200 ng)	0.132	195		
Std 7 (500 ng)	0.210	376		
R1 1	0.001	0	2.2425	0
R1.2	0.033	0	2.2619	0
R1:3	0.003	0	2.0481	0
R1.4	0.040	0	2.0562	0
R1:5	0.040	0	2.0022	0
R1:6	0.006	0	2.2757	0
R1:7	0.031	0	2.1452	0
R1.8	0.026	0	2.0308	0
R1:9	0.016	0	2.1052	0
R1 10	0.054	0	2.1445	0
R1 11	0.060	0	2.0065	0
R1 12	0.030	0	2.2509	0
R1 13	0.020	0	2.0702	0
R1 14	0.020	0	2.1444	0
R1 15	0.039	9	2.0727	10
R1 16	0.030	0	2.0608	0
R1 17	0.000	0	2.0714	0
R1 18	0.060	0	2.2542	0
R1 19	0.006	0	2.0419	0
R1:20	0.009	0	2.1244	0
R1:21	0.010	0	2.0100	0
R1:22	0.020	0	2.0015	0
R1:23	0.002	0	2.0091	0
R1.24	0.010	0	2.1306	0
R1:25	0.039	0	2.0502	0
R1:26	0.010	0	2.1701	0
R1:27	0.033	0	2.2625	0
R1:28	0.055	0.	2.1743	0
R1:29	0.007	0	2.1001	0
R1:30	0.001	0	2.3415	0
R1:31	0.004	0	2.2074	0
R1:32	0.004	0	2.1707	0
R1.33	0.000	0.	2.2705	0

Constant	0.0032
X Coef.	0.0005
Solids (g/g)	0.1011

Table C-7 (cont'd). Run 2 Reactor 1 Solids Data

Sample I.D.	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
R1:34	0.005	0	2.0556	0
R1:35	0.006	0	2.1096	0
R1:36	0.004	0	2.1730	0
R1:37	0.003	0	2.0575	0
R1:38	0.006	0	2.0690	0
R1:39	0.032	0	2.1098	0
R1:40	0.007	0	2.1175	0
R1:41	0.010	0	2.2456	0
R1:42	0.006	0	2.0530	0
R1:43	0.020	0	2.0593	0
R1:44	0.058	0	2.0996	0
R1:45	0.004	0	2.1430	0
R1:46	0.024	0	2.1707	0
R1:47	0.013	0	2.1834	0
R1:48	0.022	0	2.2002	0
R1:49	0.024	0	2.2263	0
R1:50	0.026	0	2.2540	0
R1:51	0.030	0	2.0567	0
R1:52	0.034	0	2.2770	0
R1:53	0.032	0	2.2365	0
R1:54	0.022	0	2.0860	0
R1:55	0.027	0	2.1096	0
R1:56	0.023	0	2.1515	0
R1:57	0.028	0	2.0672	0
R1:58	0.024	0	2.2480	0
R1:59	0.009	0	2.0474	0
R1:60	0.035	0	2.0675	0
R1:61	0.008	0	2.2701	0
R1:62	0.033	0	2.2610	0
	S =	0	S =	10

Table C-8. Run 2 Reactor 4 Solids Data

Sample I.D.	Absorbance (253.7)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
Blank	0.006	0		
Std 1 (30 ng)	0.027	26		
Std 2 (50 ng)	0.047	46		
Std 3 (100ng)	0.072	98		
Std 4 (200 ng)	0.124	[illegible]		
Std 5 (300 ng)	0.163	288		
R4 1	0.005	0	2.1476	0
R4:2	0.027	0	2.1775	0
R4:3	0.024	0	2.1594	0
R4:4	0.030	12	2.0454	31
R4:5	0.025	0	2.2670	0
R4:6	0.024	0	2.2637	0
R4.7	0.023	0	2.1710	0
R4.8	0.023	0	2.2611	0
R4:9	0.027	0	2.1226	0
R4.10	0.026	0	2.3264	0
R4.11	0.026	0	2.2300	0
R4 12	0.027	0	2.2153	0
R4 13	0.025	0	2.1784	0
R4 14	0.027	0	2.2008	0
R4 15	0.023	0	2.1583	0
R4 16	0.024	0	2.2694	0
R4 17	0.024	0	2.1766	0
R4 18	0.021	0	2.1200	0
R4 19	0.022	0	2.1953	0
R4 20	0.023	0	2.1711	0
R4.21	0.023	0	2.1183	0
R4.22	0.023	0	2.2010	0
R4 23	0.022	0	2.1679	0
R4 24	0.026	0	2.3480	0
R4 25	0.021	0	2.1380	0
R4 26	0.023	0	2.1562	0
R4 27	0.024	0	2.3002	0
R4.28	0.024	10	2.2480	23
R4 29	0.025	0	2.1210	0
R4:30	0.036	24	2.3190	54
R4:31	0.024	0	2.1301	0
R4:32	0.021	0	2.3090	0
R4:33	0.021	0	2.3644	0
R4:34	0.025	0	2.3006	0
R4:35	0.022	0	2.1848	0

Constant	0.0234
X Coef	0.0005
Solids (g/g).	0.1945

Table C-8 (cont'd). Run 2 Reactor 4 Solids Data

Sample I.D.	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
R4-36	0.024	0	2.1627	0
R4-37	0.022	0	2.1472	0
R4-38	0.306	0	2.1728	0
R4-39	0.023	0	2.3194	0
R4-40	0.025	0	2.1805	0
R4-41	0.023	0	2.1360	0
R4-42	0.306	0	2.2090	0
R4-43	0.024	0	2.3568	0
R4-44	0.308	8	2.3403	15
R4-45	0.024	0	2.3613	0
R4-46	0.025	0	2.1761	0
R4-47	0.026	0	2.1417	0
R4-48	0.026	0	2.1311	0
R4-49	0.027	0	2.2191	0
R4-50	0.024	0	2.1390	0
R4-51	0.028	8	2.2615	16
R4-52	0.022	0	2.4158	0
R4-53	0.025	0	2.3995	0
R4-54	0.026	0	2.3348	0
R4-55	0.024	0	2.3065	0
R4-56	0.024	0	2.3615	0
R4-57	0.023	0	2.0230	0
	S =	56	S =	148

Table C-5 Run 2 Reactor 5 Solids Data

Sample I.D.	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
Blank	0.025	0		
Std 1 (50 ng)	0.048	46		
Std 2 (100 ng)	0.072	94		
Std 3 (200ng)	0.122	194		
Std 4 (400 ng)	0.218	388		
Std 5 (600 ng)	0.318	588		
Std 6 (800 ng)	0.398	750		
R5:1	0.024	0	2.3608	0
R5:2	0.093	136	2.2458	318
R5:3	0.028	6	2.2845	13
R5:4	0.027	3	2.3485	6
R5:5	0.025	0	2.1583	0
R5:6	0.031	12	2.3042	27
R5:7	0.025	0	2.3065	0
R5:8	0.035	18	2.2030	41
R5:9	0.027	3	2.2901	6
R5:10	0.023	0	2.3412	0
R5:11	0.029	6	2.2506	14
R5:12	0.020	0	2.1522	0
R5:13	0.030	10	2.3536	22
R5:14	0.038	26	2.2378	58
R5:15	0.006	0	2.2502	0
R5:16	0.020	0	2.3163	0
R5:17	0.031	12	2.2271	28
R5:18	0.024	0	2.3318	0
R5:19	0.022	0	2.3306	0
R5:20	0.108	166	2.1828	381
R5:21	0.026	6	2.3316	13
R5:22	0.005	0	2.1930	0
R5:23	0.018	0	2.3371	0
R5:24	0.038	26	2.3455	56
R5:25	0.034	18	2.3656	40
R5:26	0.026	6	2.0918	14
R5:27	0.023	0	2.2771	0
R5:28	0.025	0	2.2266	0
R5:29	0.018	0	2.3506	0
R5:30	0.035	20	2.2202	47
R5:31	0.026	2	2.0061	4
R5:32	0.024	0	2.2433	0
R5:33	0.030	10	2.2606	22
R5:34	0.005	0	2.3316	0

Constant:	0.0256
X Coef	0.0005
Solids (g/g)	0.1984

Table C-8 (cont'd). Run 2 Reactor 5 Solids Data

Sample I.D.	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
R5:35	0.024	0	2.1666	0
R5:36	0.041	32	2.2244	75
R5:37	0.031	12	2.3476	27
R5:38	0.024	0	2.3707	0
R5:39	0.009	0	2.3279	0
R5:40	0.028	6	2.1616	14
R5:41	0.008	6	2.3727	17
R5:42	0.025	0	2.3776	0
R5:43	0.031	12	2.2677	27
R5:44	0.034	18	2.3387	40
R5:45	0.024	0	2.3154	0
R5:46	0.042	34	2.2507	79
R5:47	0.025	0	2.3248	0
R5:48	0.030	10	2.2124	26
R5:49	0.006	14	2.2672	32
R5:50	0.026	0	2.3611	0
R5:51	0.009	145	2.1898	358
R5:52	0.025	0	2.3177	0
R5:53	0.025	0	2.2378	0
R5:54	0.027	0	2.3632	0
R5:55	0.023	0	2.2391	0
R5:56	0.039	26	2.2586	45
R5:57	0.457	0	2.3763	0
R5:58	0.035	20	2.3400	45
R5:59	0.023	0	2.2920	0
R5:60	0.027	24	2.2616	45
R5:61	0.025	0	2.3005	0
	Σ =	835	Σ =	1973

Table C-10 Run 2 Reactor 8 Solids Data

Sample ID	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
Blank	0.031	0		
Std 1 (50 ng)	0.046	50		
Std 2 (100 ng)	0.058	28		
Std 3 (200 ng)	0.128	194		
Std 4 (400 ng)	0.230	408		
Std 5 (600 ng)	0.320	576		
Std 6 (800 ng)	0.420	778		
R8 1	0.073	84	2.1600	208
R8 2	0.036	0	2.2161	0
R8 3	0.034	0	2.2004	0
R8 4	0.038	16	2.3271	37
R8 5	0.130	144	2.3542	323
R8 6	0.035	8	2.3535	18
R8 7	0.032	0	2.2280	0
R8 8	0.046	28	2.3063	64
R8 9	0.042	22	2.2947	51
R8 10	0.036	10	2.1613	25
R8 11	0.086	128	2.2584	299
R8 12	0.050	20	2.2229	47
R8 13	0.036	14	2.2767	32
R8 14	0.037	12	2.2453	28
R8 15	0.128	204	2.2329	482
R8 16	0.030	0	2.2104	0
R8 17	0.030	0	2.3255	0
R8 18	0.035	8	2.1697	20
R8 19	0.048	34	2.3735	76
R8 20	0.037	12	2.2705	28
R8 21	0.035	8	2.2066	20
R8 22	0.043	24	2.2646	56
R8 23	0.047	32	2.2121	77
R8 24	0.032	0	2.3024	0
R8 25	0.032	0	2.3068	0
R8 26	0.033	0	2.2367	0
R8 27	0.030	0	2.2064	0
R8 28	0.128	188	2.3203	427
R8 29	0.050	38	2.2082	91
R8 30	0.030	0	2.2426	0
R8 31	0.040	18	2.3155	41
R8 32	0.042	22	2.1962	53
R8 33	0.045	28	2.3446	63
R8 34	0.033	0	2.2006	0

Constant:	0.0316
X Coef	0.0005
Solids (g/g):	0.1895

Table C-18 (cont'd). Run 2 Reactor 6 Solids Data

Sample I D	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
R6:35	0.027	0	2.3482	0
R6:36	0.028	0	2.2170	0
R6:37	0.032	0	2.2822	0
R6:38	0.062	100	2.1272	243
R6:39	0.038	14	2.2235	34
R6:40	0.037	12	2.3986	27
R6:41	0.072	80	2.2065	186
R6:42	0.030	0	2.3453	0
R6:43	0.035	8	2.2123	20
R6:44	0.094	106	2.2884	244
R6:45	0.033	6	2.1001	15
R6:46	0.038	14	2.2575	33
R6:47	0.041	20	2.2717	47
R6:48	0.037	12	2.3780	27
R6:49	0.030	0	2.2984	0
R6:50	0.031	0	2.3021	0
R6:51	0.078	90	2.3770	204
R6:52	0.034	0	2.3280	0
R6:53	0.031	0	2.3272	0
R6:54	0.035	6	2.3780	15
R6:55	0.037	12	2.2447	29
R6:56	0.049	36	2.2947	85
R6:57	0.047	32	2.3041	74
R6:58	0.031	0	2.2667	0
R6:59	0.087	120	2.3306	278
R6:60	0.044	26	2.3272	60
R6:61	0.030	0	2.1630	0
R6:62	0.050	32	2.1581	80
	S =		1000 S =	4984

Table C-11 Run 2 Leachate data

Sample I.D.	Absorbance (253.6)	Hg (ng)	Sample Wt. (ml)	[Hg] (ng/g)		Constant:	0.0322
						X Coef	0.0004
						Solids (g/g):	0.0052
Blank	0.033	0					
Std 1 (10 ng)	0.037	10					
Std 2 (30 ng)	0.044	29					
Std 3 (50ng)	0.052	48					
Std 4 (80 ng)	0.062	73					
Std 5 (100 ng)	0.073	100					
Std 6 (200 ng)	0.112	198					
Std 7 (300 ng)	0.153	300					
R1	0.029	0	100	0			
R2	0.025	0	100	0			
R3	0.026	0	100	0			
R4	0.029	0	100	0			
R4(dup)	0.030	0	100	0			
R5	0.020	0	100	0			
R6	0.044	28	100	53			
R7	0.027	0	100	0			
R8	0.032	0	100	0			
R8(spike)*	0.050	43	100	82		89 % recovery	
R9	0.028	0	100	0			

Table C-12 Run 2 Activated Carbon Data

Constant 0.0315
X Coef 0.0004

Sample I.D.	Absorbance (253.6)	Hg (ng)	Sample Wt. (g)	[Hg] (ng/g)
Blank	0.033	0		
Std 1 (10 ng)	0.037	10		
Std 2 (30 ng)	0.041	20		
Std 3 (50ng)	0.050	42		
Std 4 (80 ng)	0.063	75		
Std 5 (100 ng)	0.071	95		
Std 6 (200 ng)	0.112	197		
Std 7 (300 ng)	0.150	292		
ACa:1	0.026	0	4.0000	0
ACa:2	0.030	0	4.0000	0
ACa.3	0.033	0	4.0000	0
ACa 4	0.043	25	4.0000	6
ACa:5	0.046	32	4.0000	8
ACa:6	0.057	60	4.0000	15
ACa.7	0.063	75	4.0000	19
ACa.8	0.045	30	4.0000	7
ACa:9	0.071	95	4.0000	24
ACb 1	0.030	0	4.0000	0
ACb:2	0.030	0	4.0000	0
ACb.3	0.031	0	4.0000	0
ACb:4	0.033	0	4.0000	0
ACb:5	0.030	0	4.0000	0
ACb:6	0.045	39	4.9096	8
ACb.7	0.029	0	4.0000	0
ACb.8	0.035	8	4.0000	0
ACb:9	0.048	37	4.0000	9
	Σ =	383	Total	94

Table C-13. Run 2 Tygon Tubing Data

Sample ID.	Absorbance (253.6 nm)	Hg (ng)	Sample Wt (g)	[Hg] (ng/g)
Blank	0.022	0		
Std 1 (10 ng)	0.026	6		
Std 3 (30 ng)	0.035	28		
Std 4 (50 ng)	0.044	44		
Std 5 (80 ng)	0.054	64		
Std 6 (100 ng)	0.067	90		
Std 7 (200 ng)	0.112	180		
R1	0.023	0		
R2	0.022	0		
R3	0.022	0		
R4	0.031	18		
R5	0.030	16		
R6	0.036	28		
R7	0.020	-3		
R8	0.031	18		
R9	0.024	4		
	S =	98		

Constant:	0.0214
X Coef	0.0005

APPENDIX D
ABBREVIATIONS

Abbreviations

Hg = Mercury
MHg = Methylmercury
MMHg = Monomethylmercury
DMHg = Dimethylmercury
Hg^{2+} = Mercury (II)
Hg^{0} = Elemental mercury
HgS = Mercuric sulfide
S = Sulfur
C = Carbon
Fe = Iron
Cr = Chromium
Cd = Cadmium
Pb = Lead
Ni = Nickel
ZN = Zinc
CH_4 = Methane
CO_2 = Carbon dioxide
NH_3 = Ammonia
CO = Carbon monoxide
H_2S = Hydrogen sulfide
O_2 = Oxygen
N_2 = Nitrogen
H = Henry's constant
VS = Volatile solids
MSW = Municipal solid waste
SW = Solid Waste
MWC = Municipal Waste Combustion
DDDI = Double distilled deionized water
GC = Gas chromatography
CVAAS = Cold vapor atomic absorption spectrometry
CVAFS = Cold vapor atomic fluorescence spectrometry
GC-HS-MIP = Headspace gas chromatography with microwave induced plasma detection
QFAAS = Quartz furnace atomic absorption spectrometry
ICPMS = Inductively coupled plasma mass spectrometry
AED = Atomic emission detector
ECD = Electron capture detector
SBW = Slit band width
TCLP = Toxicity characteristic leaching procedure
EPA = Environmental Protection Agency

FDEP = Florida Department of Environmental Protection
CFR = Code of Federal Regulations
QA/QC = Quality assurance/quality
Cl = Chloride
OH^- = Hydroxide

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BIOGRAPHICAL SKETCH

Celia D.A. Earle was born on October 8, 1967, in Surrey England. She moved to Jamaica in 1970 and remained there until 1983. She moved to Gainesville, Florida, in 1983 and graduated from Gainesville High School in 1985. Celia immediately went to the University of Florida and received her Bachelor of Science degree in Microbiology and Cell Science in 1989. She worked as laboratory technician in a microbiology teaching laboratory from 1988 to 1989.

She worked as an assistant chemist in a water quality laboratory at the University of Florida from 1989 to 1991, and entered the graduate program in the Department of Environmental Engineering Sciences in Summer 1991. She specialized in the water chemistry area and received her Master of Science degree in August 1993. She continued on with her Ph.D. in the same department up until May 1995 when she switched to the Soil and Water Science department to complete her Ph.D. Also, in May 1995, she decided to pursue a Bachelor of Science degree in environmental engineering

She received this second Bachelor of Science degree in August 1997. She will be receiving her Ph.D. in December 1997. She is currently a member of the Society of Women Engineers, Air and Waste Management Association, American Water Works Association and numerous other societies. She intends to work in the environmental consulting field as both an environmental engineer and a scientist.

I certify that I have read this study and that in my opinion it conforms to acceptable standards of scholarly presentation and is fully adequate, in scope and quality, as a dissertation for the degree of Doctor of Philosophy.

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